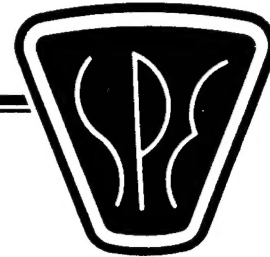


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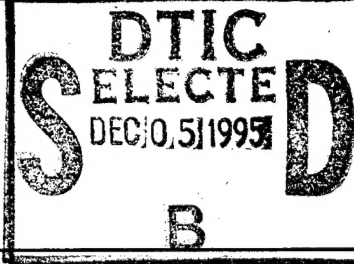
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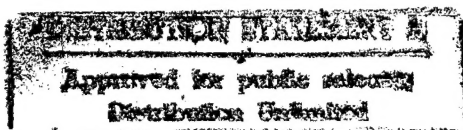
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Mr. Jack L. Isaacs, President-elect of the  
Society of Plastics Engineers, Inc., will address the  
Opening Session of the Conference.

Mr. Isaacs is affiliated with  
General Electric Company Appliance Park  
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The members of the DIVTEC Committee wish to thank  
the University of Lowell Student Chapter of the Society of Plastics Engineers  
for its significant contribution to the success of this Conference.



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## PROGRAM

Tuesday 27 September

- 8:00 Registration
- 9:00 Introductory Comments  
J.L. Isaacs SPE President Elect
- 9:15 Conference Keynote Address  
E.S. Clark University of Tennessee
- 10:00 Coffee Break
- 10:30 Apparent Modulus of a Structural Foam Member  
R.C. Progelhof New Jersey Institute of Technology  
K. Eilers Allied Chemical Company
- 11:15 Injection Molding and the Relation of Mold Filling  
Conditions to Polymer Orientation, Morphology and  
Properties  
J.L. White University of Tennessee
- 12:00 Luncheon  
Guest Speaker  
F. Reed Estabrook  
BROOK MOLDING COMPANY  
National Director S.P.I.
- 2:00 Processing for Properties - Films  
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- 2:45 Manmade Fiber Processing: Relation to Properties  
J.P. Bell University of Connecticut
- 3:30 Effect of Extrusion Processing Parameters on  
Properties and Structure of Extrudate  
N.R. Schott University of Lowell
- 4:15 Coffee Break  
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Wednesday 28 September

- 8:00 Registration
- 9:00 Blowmolding (Rheological Analysis)  
L.V. Cancio Clopay Corporation
- 9:45 Coffee Break
- 10:15 Thermoforming Processing Materials Properties and Their  
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J.E. Froehlich Moore Business Forms Inc.
- 11:00 Review of Forging, Stamping and other Solid-Phase Forming  
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K.M. Kulkarni IIT Research Institute

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- Tuesday Afternoon Professor E.S. Arnold
- Wednesday Steven Parsons, President  
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Apparent Modulus of a Structural  
Foam Member

by

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## Abstract

A theoretical analysis has been made to relate the stiffness of both rectangular and circular structural foam beams to their density profile in terms of an apparent modulus. Relationships were derived for bending, transverse compression and axial tension/compression. A secondary objective was the generation of a mathematical model in which various values of foam structural parameters, such as skin thickness, average density, density profile, etc., could be systematically substituted to yield the optimum structure from the stand point of rigidity per unit volume of material. The derived mathematical model was based upon classical strength of materials analysis. Three important assumptions were made: The density profile within the foamed core section can be approximated by a polynomial expression and the skin section is of uniform density and thickness. Further, the relationship between the local modulus of a uniform density foam section can be approximated by a density squared relationship.

The theoretical analysis and limited experimental verification clearly demonstrated the importance of its macrostructure to the rigidity of a foamed member in the prediction of beam stiffness. For a given average density the optimum properties of a member are a compromise between skin thickness, minimum core density and the shape of the density profile within the core.

For the experimental verification of the model, the density profile of the foamed member was determined by an X-ray technique. The observed density profiles were attributable to the characteristics of the plastic and to the particular molding process. Good agreement was obtained between predicted and observed values for unreinforced plastics of fine cellular structure.

## Introduction

A basic understanding of the mechanics of a structural foam member is essential if proper part utilization and mold design is to be attained. The notation "structural foam" refers to a class of thermoplastic parts possessing a high density skin and a lower density cellular core which are molded in a single operation. There are several different processes available to mold a thermoplastic structure of this nature<sup>1</sup>. These processes fall into two common categories, single component and two component<sup>1,2</sup>. In a single component process an inert gas is dispersed into the molten polymer. The polymer-gas mixture is then injected into the mold cavity. In the "low pressure" Union Carbide process the mold cavity is partially filled with the polymer-gas mixture allowing the gas to expand to fill the cavity with foam. During the expansion process the gas cells in proximity to the mold surface tend to collapse. This results in a macro-structure with a relatively high density skin and low density core. In the Dow-TAF variation, the cavity is completely filled with the polymer-gas mixture. After sufficient cooling time has elapsed to form a solid skin of finite thickness, the mold is expanded. The pressure within the molten polymer drops and gas bubbles initiate and grow to form a cellular core. The two component system uses two streams of polymers, only one of which contains gas. The two polymer streams are injected into the cavity in such a manner that a gas free polymer forms a surface skin and the polymer which contains the

gas forms a cellular core structure.

Whether the structural foam is formed by a single or two component process there exists a large density gradient across the thickness of the part. Shown in Figure 1, 2 and 3 are representative density profiles for both one and two component foam systems as measured by an X-ray transmission technique<sup>3</sup>. The mechanical properties of these polymer matrixes are dependent upon the properties of the base material and the density variation through the part. However, the density variation within the part is dependent upon the molding process, processing conditions, Mold design, blowing agent and material formulation.

If the thickness of a member could be increased while still maintaining a high density skin and constant weight, a large increase in flexural stiffness per unit weight can be obtained. However, this increase in flexural stiffness is at the expense of lower transverse compressive and axial tensile and compressive properties. Therefore this paper presents the results of an analytical and experimental investigation of the flexural, tensile and compressive properties of a single polymer structural foam member. Mathematically a structural foam member in flexure is treated differently than in simple tension or compression. Since the major applications of structural foam members are usually in flexure, this topic will be analyzed first with greater emphasis on the various analytical steps involved.

## STRESS IN BEAMS

Classical strength of materials beam analysis describing the deflection of solid polymers in flexure is based upon the following assumptions:<sup>4</sup>

- 1) The entire transverse section of the beam, originally plane, remains plane and normal to the longitudinal fibers of the bar after bending.
- 2) That the material obeys Hooke's law within the deformation limits.
- 3) That every longitudinal fiber acts as if separate from every other fiber, i.e., there are no lateral pressures nor shearing stresses between the fibers,
- 4) That the beam is initially straight and of uniform cross-section and
- 5) That the moduli of elasticity in tension and compression are equal.

Sterling<sup>5</sup> has shown that these basic assumptions are good approximations for a polymer under small strains for short periods of time, i.e. where creep is not significantly involved. Therefore, the analysis presented herein does not apply to the numerical prediction of repture or deformation under long term static levels (creep).

Under the above assumptions, the strain in the longitudinal fibers in a beam,  $\epsilon_y$ , is related to the radius of curvature of bending,  $\zeta$ , and the distance from the neutral axis,  $y$ , of the beam by:<sup>4</sup>

$$\epsilon_y = \frac{y}{\zeta} \quad (1)$$

The two equations of equilibrium expressing the net force,  $F$ , on the cross-section  $A$  is equal to zero and the bending moment is equal to the applied moment,  $M$ , are:<sup>4</sup>

$$\int_A \sigma_y dA = F_{\text{net}} = 0 \quad (2)$$

$$\int_A \sigma_y y dA = M \quad (3)$$

where  $\sigma$  is the local stress. The first equilibrium equation determines the position of the "neutral" or bending axis of the cross-section. From Hook's Law:

$$\sigma_y = E_y \epsilon_y = E_y \frac{y}{\zeta} \quad (4)$$

The moment of force of the element with respect to the neutral axis is:

$$M = \frac{1}{\zeta} \int_A E_y y^2 dA = \frac{1}{\zeta} \cdot S \quad (5)$$

where the integral 5 is commonly referred to as the "beam stiffness",  $S$ :

$$S = \int_A E_y y^2 dA$$

If the modulus,  $E_y$ , is uniform across the member as is the case of a solid polymer specimen:



$$\begin{aligned}
 M &= \frac{E_0}{\zeta} \int_A y^2 dA \\
 &= \frac{E_0}{\zeta} I_y
 \end{aligned}
 \tag{7}$$

where  $I_y$  is the moment of inertia of the section around the neutral axis. Thus, for a beam of uniform modulus the "beam stiffness" is equal to the product of flexural modulus times the moment of inertia.

In the literature the term "flexural modulus" of a structural foam member is commonly used. In this instance the beam stiffness,  $S$ , is divided by the moment of inertia of the section based upon the geometric shape of the member resulting in an apparent modulus,  $E'$ .

$$E' = \frac{\int_A E_y y^2 dA}{I_y}
 \tag{8}$$

However, this "apparent flexural modulus" is not a physical property of the material but of the system being tested which in turn is a function of the shape of the beam, polymer properties, density profile, and skin thickness. This is one of the major conclusions that this work clearly illustrates.

#### DEFLECTION OF BEAMS

Following classical strength of material analysis the curvature of a beam is related to the slope of the beam, Figure 4, by:

$$\frac{1}{\zeta} = \frac{\frac{d^2 \delta}{dx^2}}{[1 + (\frac{d\delta}{dx})^2]^{3/2}} \quad (9)$$

where  $\delta$  is the beam deflection. Assuming the slope is small compared to unity:

$$\frac{d^2 \delta}{dx^2} = \frac{1}{\zeta} \quad (10)$$

Substituting Equation 10 into Equation 5 gives the generalized beam deflection equation:

$$M = \left[ \int_A E_y y^2 dA \right] \frac{d^2 \delta}{dx^2} \quad (11)$$

To determine the deflection at a particular structural foam beam the generalized differential equation is solved for the appropriate boundary conditions. For example, a simply supported beam of uniform cross-sectional geometry with a concentrated load,  $P$ , at the center (ASTM D-790 Flexural Properties of Plastics) the maximum deflection at the center of the span given by:

$$\delta_{\max} = \frac{PL^3}{48 \left[ \int_A E_y y^2 dA \right]} = \frac{PL^3}{48S} \quad (12)$$

In the standard ASTM D-790 test the tangent to the load deflection curve,  $\tan \theta$ , at zero deflection is  $P/\delta_{\max}$ . Rearranging Equation

12 for the beam stiffness:

$$S = \frac{PL^3}{48 \delta_{\max}} = \frac{L^3 \tan \theta}{48}$$

If the modulus is uniform across the section the beam stiffness,  $S$ , is equal to  $E_o I_y$ , thus:

$$E_o = \frac{L^3 \tan \theta}{48 I_y} = \frac{L^3 \tan \theta}{4b d^3} \quad (13)$$

which corresponds to Equation 5 of section 10.11.1 of ASTM D-790 test code.

The use of Equation 13 to calculate an "apparent flexural modulus"  $E'$ , for a structural foam member is technically incorrect. However, the use of a relative "apparent flexural modulus",  $E'/E_o$ , represents the degree to which a foam polymer structure approaches the rigidity of the solid polymer of identical configuration. The relative "apparent flexural modulus" can be represented by:

$$\frac{E'}{E_o} = \frac{\int A E_y y^2 dA}{I_y E_s} \quad (14)$$

To compare structural foam reduced modulus data in flexure the exact same test, specimen size, preparation, and testing technique must be used since a material property is not being measured.

The subsequent analysis will be limited to only two basic cross-sectional geometries, rectangular and circular since these are the two most important geometries occurring in engineering design problems. In the discussion the term rectangular beam will refer

to a rectangular section cut from a plaque, that is, the density profile will be one dimensional with skin on only two opposing surfaces.

### FOAM MODULUS

To evaluate the stiffness integral for either a rectangular or circular beam the variation of local modulus,  $E_y$  or  $E_r$ , must be known. A considerable number of experimental and theoretical studies on uniform density foam have been reported in the literature. The authors have reviewed many models and data and concluded that the density approximation model:<sup>6-30</sup>

$$\frac{E}{E_0} = \left( \frac{\rho}{\rho_0} \right)^2 \quad (15)$$

appears to adequately represent the modulus density relationship for a uniform density foam having a reduced density ratio range of,  $\rho/\rho_0$  of from 0.3 to 1.0. The results of this analysis will be reported later in a separate paper. Since the relationship between modulus and density can be approximated by Equation 15 the stiffness integral,  $S$ , can be analytically determined if the variation of density across the beam is known.

### PROPOSED DENSITY VARIATION MODEL

Throne<sup>28</sup> attempted to empirically correlate the reduced density profile within a polyester structural foam plate, Figure 5, as a function of;  $R'$ , the reduced centerline density and  $C$ , a poly-

nomial curve fit parameter and non dimensional distance,  $Z$ . The relationship used was:

$$\frac{\rho}{\rho_0} = R' + (1 - R') [(C + 1)Z^C - CZ^{C+1}] \quad (16)$$

The average foam density,  $\bar{\rho}$ , of the rectangular beam was obtained by integrating Equation 16 across the cross-section:

$$\frac{\bar{\rho}}{\rho_0} = \frac{\int \rho dA}{A}$$

$$\frac{\bar{\rho}}{\rho_0} = \left[ \frac{2 + CR'}{2 + C} \right]$$

(17)

Wasserstrass and Throne<sup>29</sup> later pointed out that this model does not apply to a structural foam member with a skin of solid material of finite thickness. In this latter work the authors introduced a "shift factor" to compensate for the skin. In this paper instead of using the "shift factor" the reduced density profile has been rederived for a plate with a finite skin thickness,  $\Delta$ , as shown in Figure 6. Using Throne's notation, the reduced density profile is given by:

$$\frac{\bar{\rho}}{\rho_s} = R' + (1 - R') \left[ \frac{(C + 1)}{(1 - \epsilon)^C} Z^C - \frac{C}{(1 - \epsilon)^{C+1}} Z^{C+1} \right] \quad (18a)$$

for  $0 < Z < (1 - \epsilon)$  and

$$\frac{\bar{\rho}}{\rho_s} = 1.0 \quad (18b)$$

For  $(1 - \epsilon) < Z < 1.0$ . The average density for a rectangular beam obtained by integrating across the section is:

$$\frac{\bar{\rho}}{\rho_s} = (1 - \epsilon) \left[ \frac{2 + C R'}{2 + C} \right] + \epsilon \quad (19a)$$

Using the same form of non-dimensional density profile for a circular cross-sectional member with the parameter  $Z$  representing non-dimensional radial distance, the average density for a circular beam is:

$$\frac{\bar{\rho}}{\rho_s} = (1 - \epsilon)^2 \left[ R' + 2 (1 - R') \left[ \frac{C + 1}{C + 2} - \frac{C}{C + 3} \right] \right] + (1 + (1 - \epsilon)^2) \quad (19b)$$

Equations 18 and 19 reduce to those proposed by Throne<sup>28</sup> for a member with zero skin thickness,  $\epsilon = 0$ .

The significance of the various parameters in the density profile equation are clearly illustrated graphically in Figures 7, 8 and 9. A comparison of the results presented in these three figures clearly indicate that the average density of a structural foam member is strongly dependent upon the skin thickness, core density and polynomial shape factor. However, anyone of these three parameters can be replaced by average reduced density,  $\bar{\rho}/\rho_s$  since average density is a function of  $C$ ,  $R'$  and  $\epsilon$ , Equation 19. It is therefore expected that the physical properties of a structural foam member will also be a function of three parameters rather than the present practice, of relating properties to only

one parameter, average density,  $\bar{\rho}/\rho_s$ , Figure 10. This point can be clearly illustrated by considering the limiting cases of relative "apparent modulus" for a foamed member. The lower limit, line C in Figure 10, would correspond to a uniform density foam rectangular or circular member. The upper limit, line A for a rectangular beam, would correspond to two parallel plates of solid polymer and, line B for a circular beam, to an annulus of solid polymer.

The relative magnitude of the density profile parameters is shown by comparing the actual density profiles for four different specimens of different materials manufactured by several processes.

TABLE I  
Idealization of Structural Foam  
Density Variation

		$\epsilon$	$R'$	C
1	Polyester <sup>28</sup>	0.11	0.186	17
2	Single Component Styrene Chemical Blowing Agent	0.0625	0.45	4
3	Single Component Styrene Fluorocarbon Blowing Agent	0.2	0.44	3
4	Two Component High Density Polyethylene	0.35	0.38	5

RELATIVE "APPARENT FLEXURAL MODULUS"

A careful review of the various theoretical models found in the literature<sup>16,27,29,30</sup> for predicting the relative "apparent flexural modulus" of a structural foam member either fail to accurately model the physical system or are not based on rigorous class-



cal mechanics. It is essential to understand the relationship between relative "apparent flexural modulus" of a structural foam member and the foam macrostructure in any design optimization process. Further more, if a closed form analytical expression from which a parametric analysis can be conducted is obtainable the significance of each parameter with respect to a reduced modulus can be ascertained.

The relative "apparent flexural modulus",  $E'/E_s$ , of a structural foam member is obtained by first evaluating the "apparent flexural modulus",  $E'$ , using the density profile and the assumption that local modulus is proportional to density squared. Thus, the relative "apparent flexural modulus" of a structural foam member with respect to the skin modulus is:

$$\frac{E'}{E_s} = \frac{\int E_y y^2 dA}{E_s I_y} \quad (20)$$

For a rectangular beam:

$$\begin{aligned} \frac{E'}{E_s} &= \frac{b \int_{-h}^h E_y y^2 dy}{E_s I_y} = \frac{1}{\int_0^1 3 \frac{E_y}{E_s} Z^2 dZ} \\ &= \int_0^{1-\epsilon} 3 \left(\frac{\rho_y}{\rho}\right)^2 Z^2 dZ + \int_{1-\epsilon}^1 3 Z^2 dZ \\ &= \int_0^{1-\epsilon} 3 \{R' + (1-R') \left[\frac{C+1}{(1-\epsilon)^C} Z^C - \frac{C}{(1-\epsilon)^{C+1}}\right]\}^2 Z^2 dZ \end{aligned}$$

$$+ \int_{1-\epsilon}^1 3 Z^2 dZ$$

Evaluating the integral:

$$\begin{aligned} \frac{E'}{E_s} = 3 & \left\langle (1 - \epsilon)^3 \left\{ \frac{R'^2}{3} + 2R' (1 - R') \left[ \frac{C + 1}{C + 3} - \frac{C}{C + 4} \right] \right. \right. \\ & + (1 - R')^2 \left[ \frac{(C + 1)^2}{2C + 3} - \frac{2C (C + 1)}{2C + 4} + \frac{C^2}{2C + 5} \right] \} \\ & \left. + \frac{1}{3} [1 - (1 - \epsilon)^3] \right\rangle \end{aligned} \quad (21)$$

For a section with a uniform foamed core,  $C = \infty$  it reduces to 8:<sup>27</sup>

$$\frac{E'}{E_s} = (1 - \epsilon)^3 R'^2 + [1 - (1 - \epsilon)^3] \quad (22)$$

And for a circular beam:

$$\begin{aligned} \frac{E}{E_s} = 4 & \left\langle (1 - \epsilon)^4 \left\{ \frac{R'^2}{4} + 2R' (1 - R') \left[ \frac{C + 1}{C + 4} - \frac{C}{C + 5} \right] \right. \right. \\ & + (1 - R')^2 \left[ \frac{(C + 1)^2}{2C + 4} - \frac{2C (C + 1)}{2C + 5} + \frac{C^2}{2C + 6} \right] \} \\ & \left. + \frac{1}{4} [1 - (1 - \epsilon)^4] \right\rangle \end{aligned} \quad (23)$$

with a uniform core,  $C = \infty$  it reduces to:<sup>27</sup>

$$\frac{E'}{E_s} = (1 - \epsilon)^4 R'^2 + (1 - (1 - \epsilon)^4) \quad (24)$$

The effect of the various foam density parameters have on the relative "apparent flexural modulus",  $E'/E_s$ , can now be determined from the above analytical equations.

Equations 21 and 23 for relative "apparent flexural modulus" for both rectangular and cylindrical beams in flexure were solved on the digital computer as a function of  $C$  and  $R'$  for a different levels of relative skin thickness,  $\epsilon$ . The results indicate that the relative "apparent flexural modulus" of a beam with a low core density is strongly dependent upon skin thickness. However, a more important graphical comparison can be made by using reduced density,  $\rho/\rho_s$ , as the independent parameter. The data for  $C = 3$ , 5 and  $\infty$  has been plotted in Figures 11, 12, and 13 respectively. For a particular value of shape factor,  $C$ , the difference in relative "apparent flexural modulus" between a rectangular and circular beam is small. Plotting relative "apparent flexural modulus"  $E/E_s$  versus skin thickness,  $\epsilon$ , for a rectangular beam, Figures 14 and 15 indicates the possible variation in relative "apparent flexural modulus" for a constant density specimen with different density profiles. The variation of relative "apparent flexural modulus" with shape factor is large.

The practical significance of these variations can be illustrated in showing the variation in relative "apparent flexural modulus" as a function of the distance from the sprue. Experimentally it has been shown that the local average density of a structural foam part is relatively insensitive to distance from the sprue. However, an examination of the cross-sectional density profile indicates that there is a significant variation of profile with flow length. Near the sprue the skin thickness is relatively large and the core density relatively uniform whereas at the extremities the skin approaches zero thickness and the

density profile is much more gradual. The magnitude of the variation in cross-sectional properties can be clearly illustrated using an average part density of 60%: From Figure 15:

<u>Position</u>	<u><math>\epsilon</math></u>	<u>C</u>	<u><math>E'/E_s</math></u>
Near Sprue	0.35	$\infty$	0.62
Near Extremity	0.0	3	0.73

This represents a variation of reduced flexural modulus of approximately 17% along the flow path length. Similar results are also true for higher average part densities and for a circular beam. If the average density of a part varies significantly with flow length the deviation can be much larger.

#### STIFFNESS TO WEIGHT RATIO IN FLEXURE

In addition to the relative stiffness of a beam the stiffness to weight ratio indicates how economically efficient the polymer in the structural foam is being used in a load bearing situation as compared to a solid beam. A measure of this efficiency is the relative flexural load bearing factor, FLBF, for two beams of identical thickness, is defined as:

$$FLBF = \frac{\frac{E'}{E_s}}{\frac{\bar{\rho}}{\rho_s}} = \frac{\frac{E'}{\bar{\rho}}}{\frac{E_s}{\rho_s}} \quad (27)$$

A comparison of the "stiffness to weight" ratio can also be made on an equal weight basis per unit width. However, the thickness of the foamed beam will be significantly greater than the solid beam. This approach is not presented here but will be presented in a subsequent paper.

Due to the complexity of the mathematics there is no simple technique for finding the maximum FLBF as a function of the structural foam parameters. Numerical results for both rectangular and circular beams were obtained on the digital computer and graphically presented.

Shown in Figures 16 through 18 are plots of FLBF for a rectangular and circular beam as a function of shape factor  $C$  and skin thickness  $\epsilon$ , for constant core density ratios,  $R'$ , of 0.2, 0.4 and 0.6. The  $R'$  parameter was chosen as an independent variable since it is the authors contention that one of the limiting design conditions for a structural foam member will be the transverse strength of the rectangular beam. The shape of the curves indicate that FLBF is a strong function of skin thickness and core density ratio. However, above a relative skin thickness,  $\epsilon$ , of 20% the shape factor,  $C$ , has relatively small effect on FLBF for the core densities examined. This fact indicates that in a structural thickness of from 20 to 40%. Not only is the FLBF near a maximum over a large range of shape factors,  $C$ , but the relative "apparent flexural modulus", Figures 11, 12, and 13 is also high. The data was cross-plotted for a rectangular beam Figure 19 to show the relationship between  $C$  and  $R'$ , for constant  $\epsilon$  which gave the maximum FLBF ratio. Superimposed on the graph as dotted lines are iso-maximum values to indicate the direction for maximum FLBF ratio. The curves indicate that maximum FLBF occurs at  $R \rightarrow 0$  at  $C \rightarrow \infty$ . However, from a practical viewpoint this is impossible. However, core densities of 20% and skin thickness of 20% at  $C \rightarrow \infty$  might be obtainable which are in the direction of maximum utilization of polymer from a

flexural stiffness to weight ratio. The results obtained for a cylindrical beam exhibit similar characteristics to that of a rectangular beam but the maximum FLBF is below that of a comparable rectangular section.

### "APPARENT MODULUS" IN AXIAL TENSION OR COMPRESSION

In either an axial tension or compression mode it is assumed in classical strength of materials analysis that the deformation across the entire transverse section is uniform. Thus, the total load carried by the member is:

$$F = \int_A \sigma \, dA = \int_A \epsilon \, E_y \, dA \quad (28)$$

Again using the notation of a relative "apparent T/C modulus" of the section:

$$\frac{E'}{E_s} = \frac{\int_A E_y \, dA}{A \, E_s} \quad (29)$$

evaluating the integral for a rectangular beam:

$$\begin{aligned} \frac{E'}{E_s} = (1 - \epsilon) & \left\langle R^2 + \frac{4R(1 - R)}{(C + 2)} + (1 - R)^2 \right. \\ & \left. \left[ \frac{(C+1)^2}{2C+1} - C + \frac{C^2}{2C+3} \right] \right\rangle + \epsilon \end{aligned} \quad (30)$$

and for a circular beam:

$$\begin{aligned} \frac{E'}{E_s} = (1 - \epsilon)^2 & \left\langle R^2 + 4R \frac{(1 - R)}{(C + 3)} \frac{(2C + 3)}{(C + 2)} \right. \\ & + 2(1 - R)^2 \left[ \frac{C + 1}{2} - \frac{2C(C + 1)}{2C + 3} + \frac{C^2}{2(C + 2)} \right] \\ & \left. + [1 - (1 - \epsilon)^2] \right\rangle \end{aligned} \quad (31)$$

Equations 30 and 31 for relative "apparent T/C modulus" were solved on the computer. Numerical results for both rectangular and circular geometries for  $C = 3, 5$  and  $\infty$  has been plotted in Figures 20, 21 and 22 respectively. The data was cross plotted, Figure 23, to show the effect of skin thickness and shape factor on reduced "apparent T/C modulus" for a constant density. A comparison with the results in flexure, Figure 14, clearly indicates that reduced axial and tensile "apparent modulus" for a fixed density is not as sensitive to skin thickness as flexural properties. Using the same parameters as in the previous flexural example:

<u>Position</u>	<u><math>\epsilon</math></u>	<u>C</u>	<u><math>E'/E_s</math></u>
Near Sprue	0.2	$\infty$	0.40
Near Extremity	0.0	3	0.41

#### STIFFNESS TO WEIGHT RATIO IN AXIAL TENSION/COMPRESSION

In a similar manner the relative stiffness of a structural foam member in axial tensile/compressive, TCLBF, can be evaluated as:



$$TCLBF = \frac{\frac{E'}{E_s}}{\frac{\bar{\rho}}{\rho_s}} = \frac{\frac{E'}{\bar{\rho}}}{\frac{E_s}{\rho_s}} \quad (32)$$

Plots of TCLBF for fixed core density ratio,  $R'$ , of 0.2, 0.4 and 0.6 are shown in Figures 24, 25, and 26. As can be seen the increase in flexural properties exhibited in the previous graphs is obtained at the expense of the tensile/compressive load bearing capability of a structural foam member. The figures clearly indicate that for a given core density the skin thickness is a highly sensitive parameter. A member with a small skin thickness; for example,  $C = 5$ ,  $\epsilon = 0$ ,  $R' = 0.2$ , shows a loss of approximately 40% of its tensile/compressive properties as compared to a solid member. However, the curves plotted as  $E'/E_s$  vs  $\bar{\rho}/\rho_s$  and TCLBF vs  $C$  indicate that the greater the skin thickness the lower the loss of tensile/compressive properties for a particular value of  $C$  or  $\bar{\rho}/\rho_s$ .

#### "APPARENT MODULUS" IN TRANSVERSE TENSION OR COMPRESSION

The transverse direction is perpendicular to the skins of a rectangular beam. It is commonly referred to as the "crushing" direction of a foam sheet. In either a transverse tension or compression mode it is assumed in classical strength of materials analysis that the stress at any section is the same. Thus, the total deflection of the body is the:

$$\delta = \int_{-h}^h d\delta = \int_{-h}^h \epsilon dy \quad (33)$$

Again assuming that the material obeys Hooke's law and that local modulus is related to density squared:

$$\delta = 2 \int_0^h \frac{\sigma}{E_y} dy = \frac{2\sigma h}{E_s} \int_0^1 \frac{dZ}{\left(\frac{\rho}{\rho_s}\right)^2} \quad (34)$$

For a solid member the displacement is:

$$\delta_s = \frac{2\sigma h}{E_s} \quad (35)$$

Thus, a reduced transverse tensile/compression modulus for a structural foam member can be defined as:

$$\frac{E'}{E_s} = \frac{1}{\int_0^1 \frac{dZ}{\left(\frac{\rho}{\rho_s}\right)^2}} \quad (36)$$

Using the density profile model, Equation 36, becomes:

$$\frac{E'}{E_s} = \frac{1}{\int_0^1 \frac{dZ}{\left\{ R' + (1-R') \left[ \frac{(C+1)}{(1-\epsilon)^C} Z^C - \frac{C}{(1-\epsilon)^{C+1}} Z^{C+1} \right] \right\}^2} + \epsilon} \quad (37)$$

This equation cannot be readily integrated in closed form but a numerical technique<sup>32</sup> based upon a 16 point Gauss-Legendre

quadrature was developed. The numerical approximation was verified by using a 100 step Romberg integration algorithm at several representative values of  $R$ ,  $C$  and  $\epsilon$ .

Shown in Figure 27 is a plot of reduced modulus as a function of the profile shape factor for different reduced core densities. The reduced modulus is strongly dependent upon profile shape factor,  $C$ , but from a design point the limiting factor is the reduced density. This can be more clearly seen in Figure 28. For a reduced density of 0.6 and appreciable skin thickness,  $0.2 < C < 0.4$  the reduced modulus is very small. This can be explained in that under these conditions the reduced core density ratio,  $R'$ , is approaching zero. At a higher average density, 0.8, the effect is not as profound since higher core densities are present.

TABLE 2  
Comparison of Results

Material/Process	Density Parameters			Flexure		Tension/ Comparison		Transverse Compress	
	$\epsilon$	R	C	Theory	Exp.	Theory	Exp.	Theory	Exp.
1. Single Component Styrene* Chemical Blowing Agent	0	.48	5	.64	.75	.43	.41		
2. Single Component Styrene Fluorocarbon Blowing Agent	.2	.44	3	.86	.89	.58	.46		
3. Two Component High Density Polyethylene	.35	.38	5	.85	.85	.58	.53	.30	.21
4. Two Component Polypropylene -Expanding Mold	.10	.45	$\infty$	.42	.46	.28	.28	.19	.15

\*Surface Modulus equal to  $2.5 \times 10^5$  psi

### Experimental Verification

In the preceeding section it was shown how the theoretical "apparent modulus" of a structural foam member can be calculated as a function of the density profile within the section and the base resin properties. To verify this work a 25 MM wide flexural bar, ASTM type I tensile bar, 50 square MM compression square and a 50 MM long density profile specimen bar were cut from the same location on a large single plaque. Thus, the four test specimens represented a rectangle of approximately 100 by 250 MM. However, the actual area in which the apparent modulus properties were measured is only about 100 by 100 MM. It was assumed that throughout this area the density profile is relatively uniform. Each plaque was carefully examined by light transmission techniques to eliminate cutting from a nonhomogeneous section, although the authors realize that it was impossible to obtain totally uniform test sections conditions in all the test specimens. A visual examination often revealed nonuniform cell structure in at least one of the four specimens. Thus, there will be some expected deviation of the experimental results from the predicted values. A comparison of both results for four different polymer/processing conditions is presented in Table 2.

The comparison of apparent moduli based upon solid resin properties for the single component fluorocarbon blown styrene and both two component specimens is very good. However, the single component GPS styrene experimental results did not correlate at all with the theoretical results when the properties of

the foam skin were assumed to be that of unfoamed GPS,  
 $E = 4 \times 10^5$  psi. This anomaly resulted in retesting with new  
tensile and flexural specimens. However, the new results were  
equivalent to the first series of experimental results. A  
further investigation of the foamed specimen indicated that the  
microbubble structure was prevalent at the surface of the speci-  
men. Several thin slivers were carefully cut from the surface  
of the specimens. Tensile modulus for these specimens averaged  
about  $2.5 \times 10^5$  psi. In addition local density of the skin  
varied from 0.90 to 1.00 depending upon location. A correlation  
of relative "apparent moduli" using the lower tensile modulus  
values were much better agreement with the predicted values.

## Conclusions

The theoretical model presented in this work indicates that to maximize the "stiffness" to weight ratio of a structural foam member the member should have a minimum skin thickness of 20%. At this physical condition the loss in axial relative "apparent modulus" is minimized. The "apparent" transverse modulus has been shown to be strongly dependent upon the centerline core density,  $R'$ . Thus, for a particular structural foam member an optimum density profile would be a relatively uniform center section with a finite transition area from the core density to the skin,  $C \approx 10$ , and a skin of approximately 20 to 30% of the total thickness. The minimum core density is determined by the transverse loading conditions.

The theoretical model as presented in this work does not include the effect of bubble size and random voids which are more important. However, the authors believe that through additional experimental data a reliability criteria or correction factors can be developed. When using the theoretical model to predict the apparent modulus of a single component structural foam member the modulus of the skin must be known. Our investigation clearly showed that the properties of the skin are not necessarily equal to that of the unfoamed polymer.

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## NOMENCLATURE

A	Cross-sectional area
b	Beam width
C	Density profile shape factor
d	Beam depth: $2h$
D	Diameter
E	Modulus
F	Force
h	Beam half thickness
I	Moment of inertia
L	Beam span
M	Moment in beam
P	Load
R	Radius of circular beam
r	Distance from centerline
R'	Nondimensional core density: $\frac{\rho_{cl}}{\rho_s}$
S	Beam stiffness
x	Distance along neutral axis

y	Distance from neutral axis
Z	Nondimensional distance: $\frac{y}{h}$ or $\frac{r}{R_0}$
$\Delta$	Skin thickness
$\delta$	Deflection
$\rho$	Density
$\bar{\rho}$	Average density
$\sigma$	Stress
$\zeta$	Radius of curvature of a loaded beam
$\epsilon$	Strain
$\epsilon$	Nondimensional skin thickness: $\frac{\Delta}{h}$

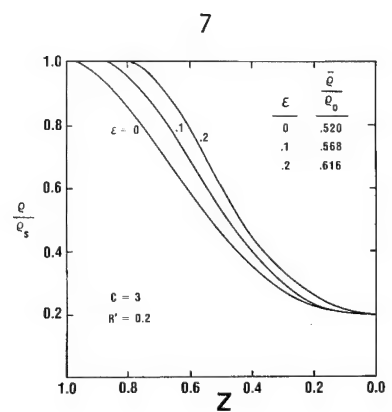
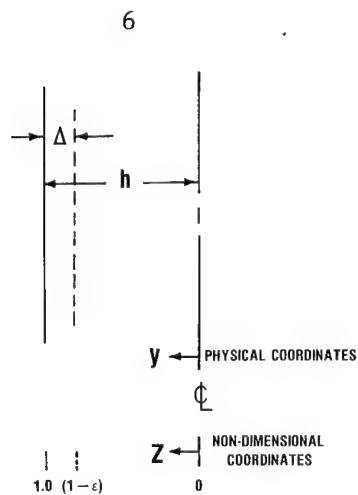
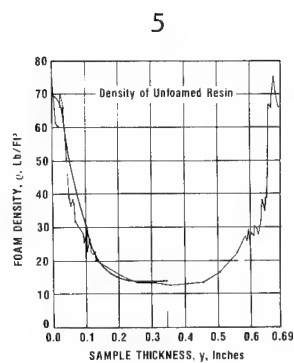
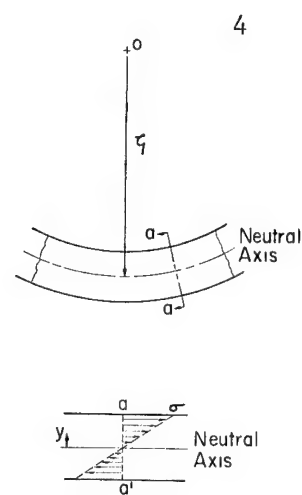
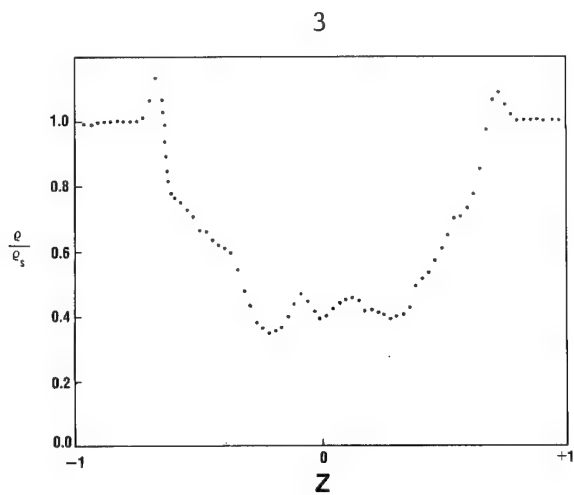
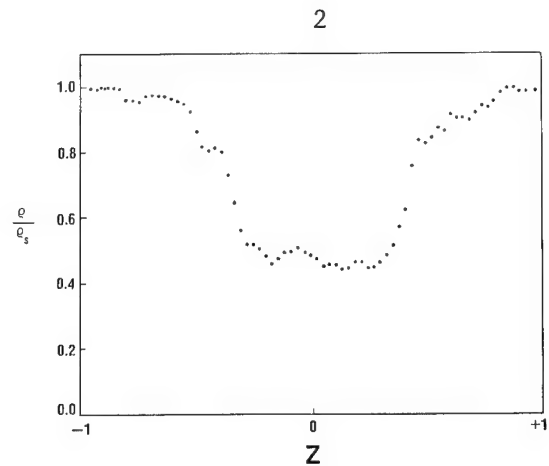
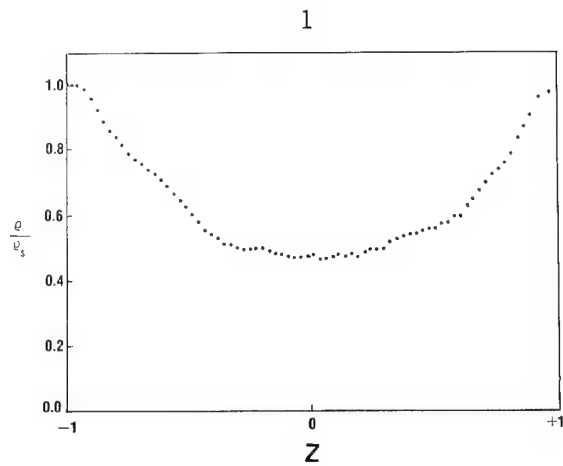
#### Subscripts

sf	Structural foam
o	Unfoamed resin
s	Surface
cl	Center line

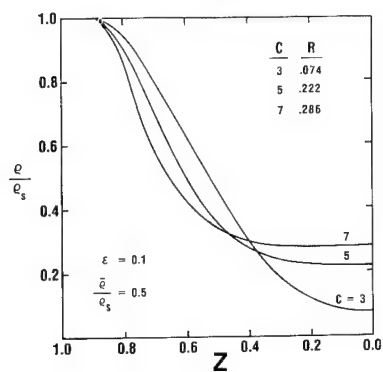
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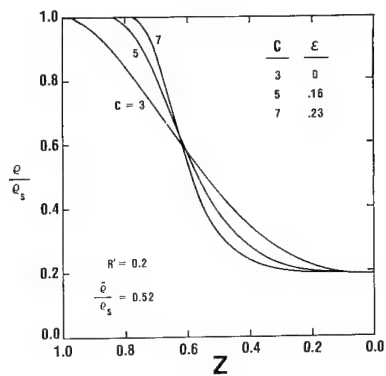
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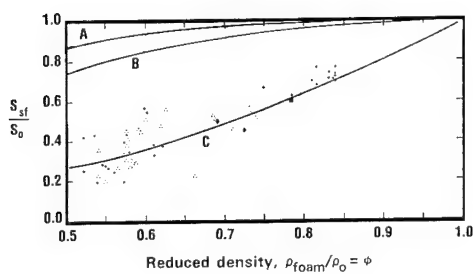
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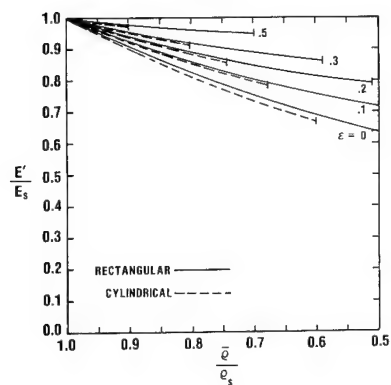
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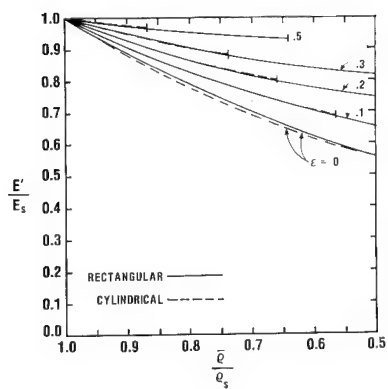
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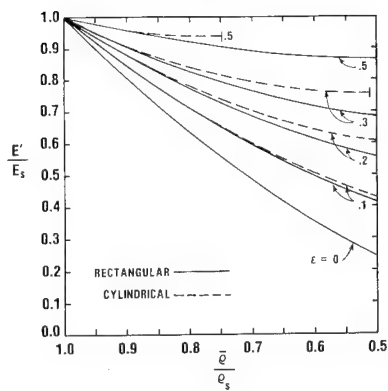
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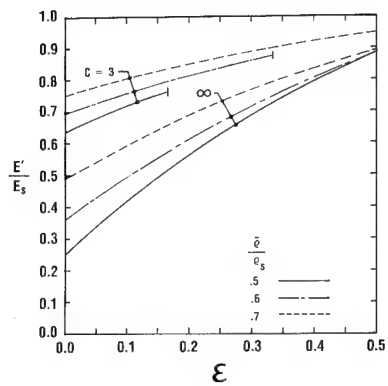
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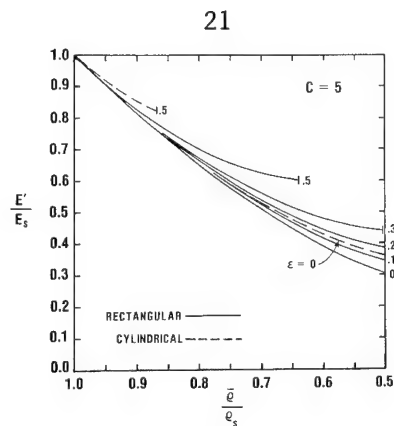
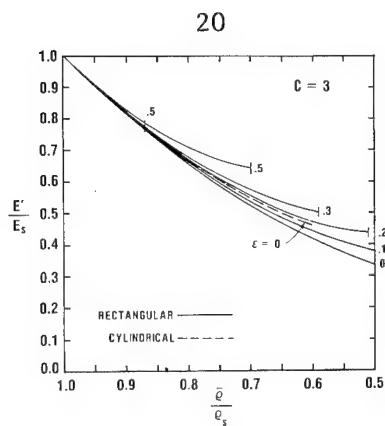
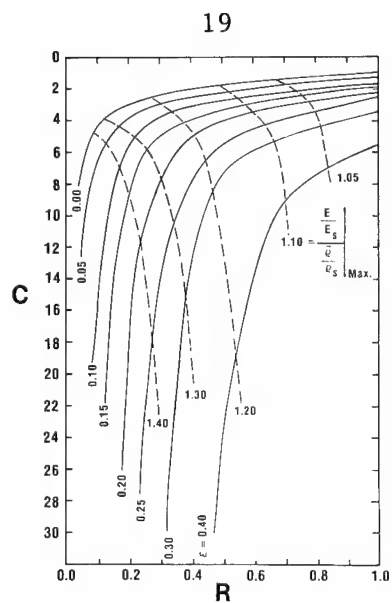
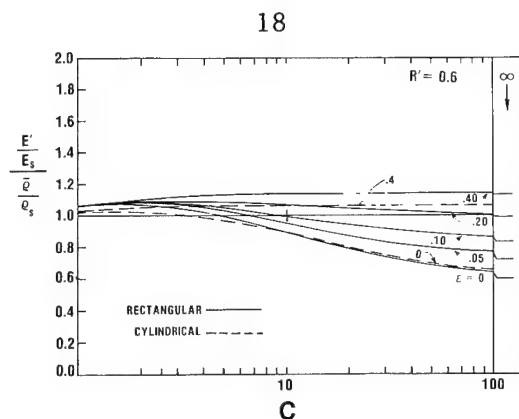
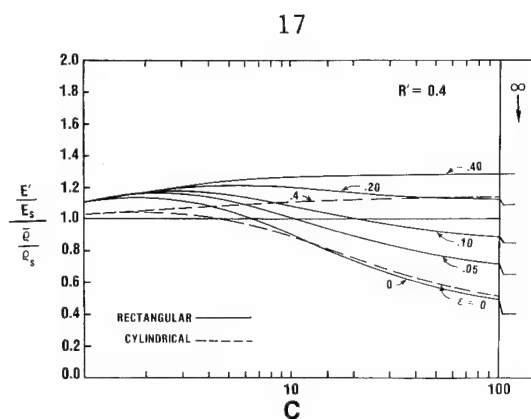
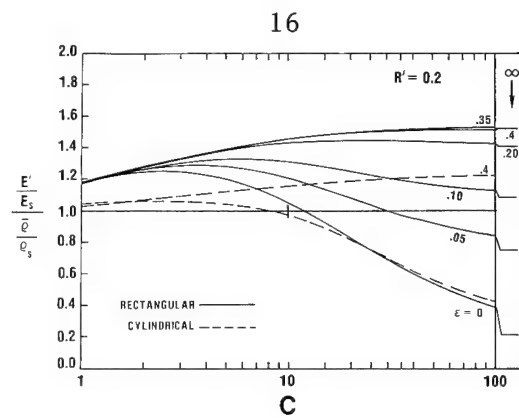
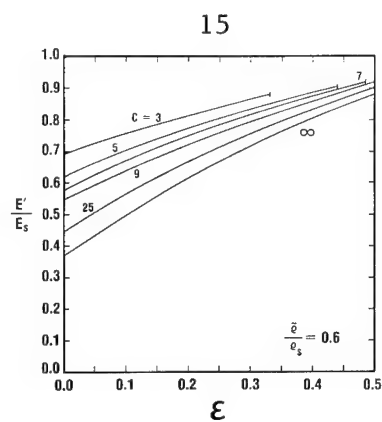


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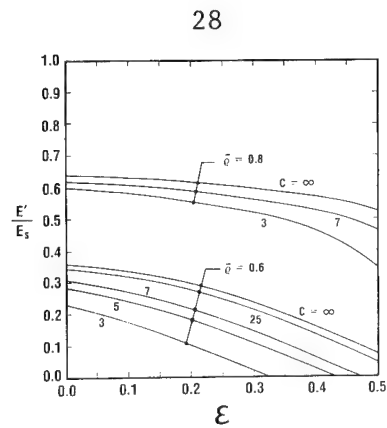
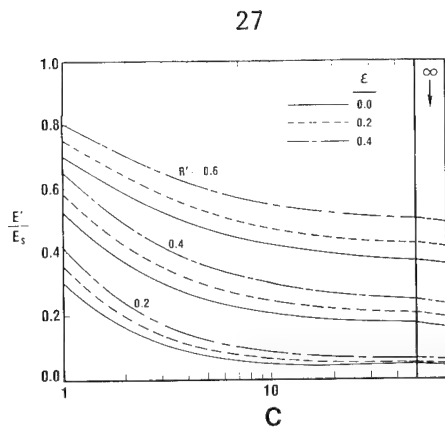
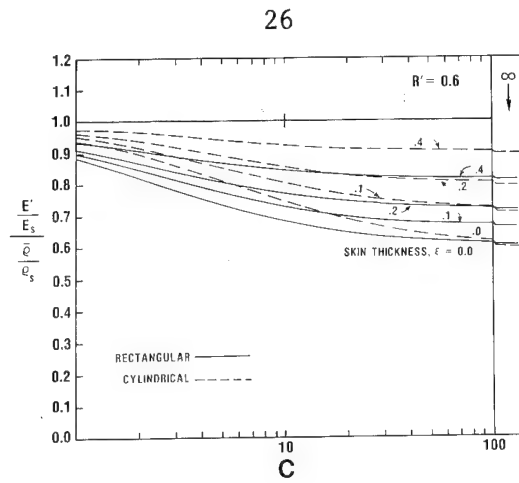
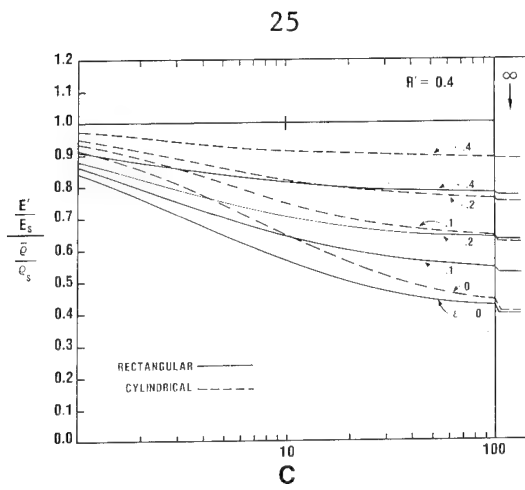
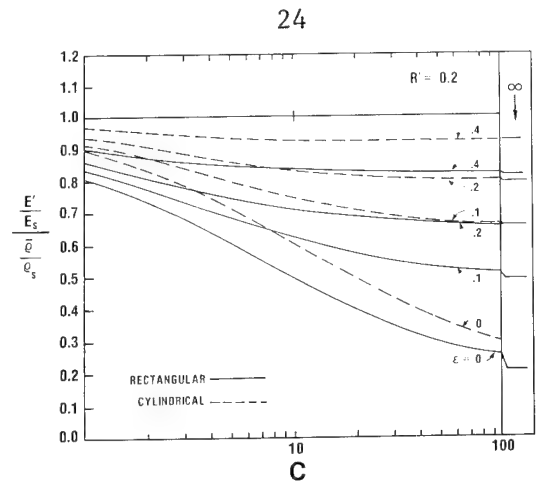
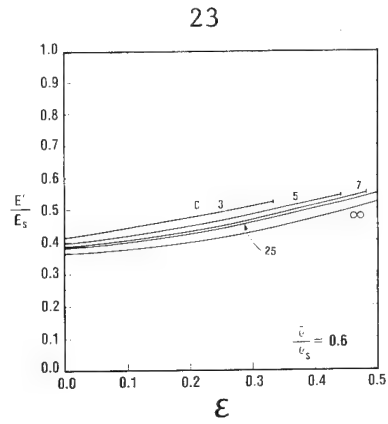
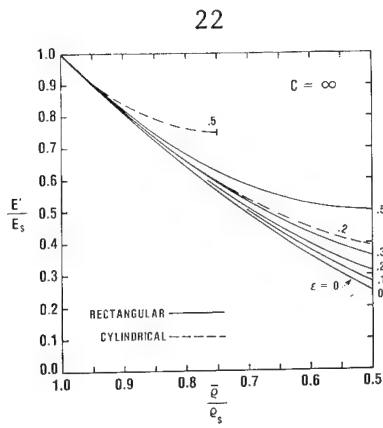


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INJECTION MOLDING AND THE RELATION OF MOLD FILLING  
CONDITIONS TO POLYMER ORIENTATION,  
MORPHOLOGY AND PROPERTIES

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## INTRODUCTION

No polymer processing operation is more important than injection molding in the production of discrete fabricated products and structural components. It has long been known that the detailed conditions of molding can strongly influence the structure and properties of molded parts. However, the situation has also been realized to be a complex one depending on injection rate, mold temperature, gate location and design and other problems. In this paper we discuss the flow phenomena occurring during injection molding and their relationship to the development of orientation and mechanical properties.

## MOLD FLOW CONDITIONS

It has been observed by various investigators (1,2,3) that there are two regimes of mold filling, one in which there is an expanding front which completely fills the mold and a second in which the melt issues as a jet and shoots across the mold. The existence of these two regimes greatly complicates the question of the influence of molding conditions on orientation and morphology in molded parts. The jetting regime is the less common but more troublesome. Here the melt surges in as a jet until it strikes a barrier. The jet then piles up on itself and eventually melt from the gate fills the mold. In a cold mold, heat will be withdrawn from the jet and it will begin to solidify. The melt which now comes in and fills the mold does not have the heat content to re-melt and fuse with the jet, the resulting molded part will not only have surface blemishes but have weld lines. Such weld lines will act as defects and may well seriously deteriorate the mechanical properties of molded parts.

Mold filling by a developing front is a more regular motion and more subject to detailed mathematical analysis. Many investigators (4,5,6,7) have developed theoretical analyses of the non-isothermal fluid mechanics of mold filling in this regime generally for disc or plate shaped molds. The analyses can essentially be considered as being based on hydrodynamic lubrication theory. Some authors (8,9,10,11) have noted that under some conditions Hele-Shaw flow theory is applicable and that the front moves through the mold in such a manner that the streamlines (when the mold is viewed parallel to the narrow section) are the same as those given by potential flow theory. Viewed perpendicular to the narrow cross-section one has solidified layers on the mold walls and an approximately isothermal hot core through which the melt moves with an approximately "power law" type velocity profile. At the front the melt from the center region flows outward like a fountain to the mold walls where it solidifies. This type of flow continues with the injection rate being determined from the ram pressure by hydrodynamic considerations.

The criterion of which of the two regimes occurs has been investigated by Oda, White and Clark (3). They find that the transition is determined by the magnitude of the extrudate swell of the melt emerging from the gate. If this is large enough to contact the mold walls, then the developing front regime occurs. If this is not the case, jetting occurs. The problem is more complicated than the simple "die swell" problem because the melt entering the cold mold through a cold sprue and runner is often of unknown temperature and, indeed, of temperature which varies with injection rate because of heat transfer.

## ORIENTATION IN AMORPHOUS INJECTION MOLDED PARTS

Orientation in injection molded parts of amorphous plastics has been studied since the work of Gilmore and Spencer (11). Quantitative studies of orientation variations in parts molded in narrow channels were carried out by Ballman and Toor (12) and by later researchers (13-16). Menges and Wubken (17,18) and Machholz (19) have studied this problem by slicing small samples from injection molded parts and determining their shrinkage. They equate their shrinkage distributions to orientation distributions.

Generally it is found that orientation is a minimum in the center of simple injection molded parts and when plotted as a function of position in thin walled parts exhibits a maximum at an intermediate position near the outer surfaces. In some studies a second maximum at the outer surface is found and often shoulders between the first maximum and the core. Generally the orientation is higher nearer the gate than further along the mold. Studies of the variation of orientation distributions with molding conditions have shown that the major processing variables appear to be injection pressure with orientation levels increasing with increasing pressure.

A mechanism to explain orientation distributions in injection molded parts has been proposed by Ballman and Toor (12) and developed by Tadmor (20) and by Dietz, White and Clark (21). The mold is envisaged to have during filling a partially solidified layer of thickness  $\Delta$  along the mold wall. The orientation maxima are attributed to the high shear stresses induced by melt surging along this layer. The motion

of melt at the front plays a key role in determining orientations near the surface of parts. A fountain-like flow at the front deposits melt on the cold walls where it solidifies. The kinematics of the front determine orientation levels in this material.

Dietz, White and Clark (21) use the methods of rheo-optics to predict orientation distributions across the narrow cross-section in injection molded parts. They use in this analysis the result of Oda, White and Clark (22) that rheo-optics may be applied to non-isothermal deformations and the birefringence existing at the glass transition temperature is frozen in. In effect they take the principal rates of the stress and refractive index tensors to be coaxial above the glass transition temperature. The differences in the magnitudes of this principal axis and magnitudes of the two tensors are presumed proportional. They are able to predict the generally observed birefringence distributions in injection molded parts with intermediate maxima as described above. Quantitative agreement is shown between the theory and selected data. As the difference in principal stresses in shear flow is uniquely related to the shear stress which is proportional to the pressure gradient, injection pressures are predicted to be the primary variable in inducing orientation.

Birefringence distributions for more complex injection molded parts are shown by Koita (23) and by Cleereman (24). Koita was concerned with the molding of telephone housings and Cleereman with molding annular parts using a mold with a rotating core. It is also possible to predict orientation distributions in complex moldings of this type (25).

## MORPHOLOGY OF CRYSTALLINE INJECTION MOLDED PARTS

The pioneering studies of the character of crystalline injection molded parts are due to Clark (26-29). Other investigators have carried out similar studies in recent years (30-32). Clark has proceeded by making polarized light optical photomicrographs of the cross-section and transmission electron micrographs of the surfaces of molded bars. A randomly oriented spherulitic core was found together with increasing orientation as one proceeds to the surface of the molded part. The surfaces of the molded parts appear to exhibit "row structures" (33). This view is generally confirmed by other investigators. Clark has considered variations in such morphologies brought about by changes in molding conditions and has shown how in some cases these lead to defective molded parts (29). He notes, for example, variations induced by changes in the screw forward time.

These studies are largely of a qualitative character. There is also no general analytical procedure for predicting crystalline morphology formed in complex deformations. A start in this direction has been made for uniaxial deformations by Spruiell and White and their coworkers (34,35) who have related crystallization kinetics to a combination of cooling rates and applied stresses and orientation development in crystalline polymers to applied stresses in the melt.

## RESIDUAL STRESSES

If portions of a molded part are constrained during cooling, density variations caused by cooling act as strains and induce stresses, such stresses known as thermal or residual stresses. This type of stress

development has long been realized and is surveyed, for example, in the old monograph of Timoshenko and Goodier (36). Its importance in plastics has been described by various investigators (19,37-41) and considered in studies of quenched sheets and injection molded parts. Methods of calculation of residual stress levels in injection molded parts have been discussed by Knappe (37) and Targiel (41).

#### MECHANICAL PROPERTIES

Structural and orientation distributions as well as residual stresses in injection molded parts induce variations in mechanical properties. Studies of the influence of molding conditions on injection molded parts have been carried out in numerous research groups. The results vary with mold design due to the interaction of heat transfer and flow in inducing stresses and the possible occurrence of jetting (3) which gives rise to surface blemishes, weld lines and undoubtedly mechanical defects (42). If we can neglect the jetting regime and weld lines in general, then mechanical properties will correlate with orientation. Modulus and tensile strength increase with increasing orientation levels. As high injection pressures produce high orientations, they will lead to higher modulus and tensile strength. An extensive study of this type is given by Machholz (19).

It must be realized that injection molded parts are structurally and mechanically heterogeneous. This is nicely shown, for example, by Heckmann and Johnson (32) who measure the stress-strain curves of thin sections removed from various positions in the cross-section.

Improper molding which leads to peculiar morphologies and residual stresses can lead to early performance. However, unique designs of



molding procedures such as Cleereman's rotating core mold (24) can lead to improved properties because of the unique orientation distribution.

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PROCESSING FOR PROPERTIES - FILMS

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We use synthetic polymer films in one form or another almost every day. At breakfast, the bread or the muffin is wrapped in a plastic film. Often the morning paper is protected from inclement weather in a polyethylene film wrapper. The ever-present sandwich bag and the roll of food wrap have become common household items. In fact, these kinds of polymer films have become so much a part of our lives that we become unaware they are around. Nevertheless, the film business is a significant part of our economy. In 1975 there were about 2500 extrusion lines of various kinds in the United States which produced about 5.5 billion pounds of self-supported plastic film<sup>1,2</sup>. It was reported recently that plastics used in packaging applications will double by 1985, to about 11.5 billion pounds<sup>3</sup>. We should define a plastic film as a planar sheet, thick enough to be self-supporting and thin enough to be flexed or creased without cracking. To be more specific we will be discussing films that range in thickness from roughly 0.1 mil to 15 mils.

Although there are many film forming polymers, we can expect that no more than 20 will enjoy and maintain continued commercial success. The major factor is cost. Few polymers with a price in excess of \$2/lb. are fabricated into films. The economic driving force resides in a concept called "value-in-use". We can think about this in the following way. For every societal need, there usually are several alternative products that can fulfill the need requirements at least to some degree. A particular product may not

be the best; but sufficient may be good enough. Accordingly, the plastics engineer is ultimately confronted with generating some acceptable balance between material costs, processability and useful properties. The latter two, processing and properties, are the topics of interest at this meeting.

Polymeric films are used in a variety of ways. Packaging uses are fairly obvious, but there are also many industrial uses. A partial listing of the functions that we expect from films is shown in Figure 1. Films are used to enclose or encapsulate solids, liquids or vapors. They serve as a carrier, e.g. of a photographic emulsion, to store an image or, of a magnetic domain to store a sound "image". The film may become a dielectric component of a capacitor, or an engineering material imparting strength or rigidity to some structure. We also require that several of these functions operate simultaneously and cooperatively. For example, a magnetic tape requires high strength/unit cross sectional area, good adhesion of the magnetic coating to the film base and a surface relatively resistant to abrasion and static. Accordingly, the two general areas of properties that are important to film technology are:

1. Bulk Properties
2. Surface Properties.

We can illustrate and emphasize this duality best with concrete examples of two important, but diverse film uses: microfilm and capacitor tissue, Figures 2,3. Materials for both uses can be made from the same basic polymer by the judicious application of processing technology.

The polymer used to make a film determines in large measure the end result. Bulk properties are developed by the bonding characteristics and the molecular symmetry of the polymer chain. Of primary importance is the molecular weight. The effect of molecular weight on properties is frequently generalized into the type of curve shown in Figure 4. At very low molecular weights polymers tend to be brittle and friable. As the molecular weight is increased many properties begin to improve rapidly once some critical average degree of polymerization  $\overline{DP}_c$  is obtained.  $\overline{DP}$  is a measure of the average number of repeat units in the polymeric chain. For poly(ethylene terephthalate) the  $\overline{DP}_c$  is 25 and is equivalent to a molecular weight of about 5000. Properties then increase rapidly until a point (K) is reached where the rate of improvement starts to decrease. For economic and processing ease, polymers are usually made to the lowest molecular weight that will generate the properties required. This is slightly above the knee of the curve (K). With some properties, additional increases in molecular weight add very little improvement and we reach a point of diminishing returns. However, other properties of interest may be found to increase again at very high molecular weights, and a curve with two plateaus occurs, as shown in Figure 5.

Chemical constitution of the polymer determines to a large degree the thermal and electrical properties of films. Figure 6 compares the thermal characteristics of several different film forming polymers. Probably the most significant term is the "upper limit service temperature" which is the maximum temperature that should be considered for extended periods. Zero strength temperature is that temperature where the film can no longer support its own weight.

Once the polymer choice has been made, the next problem is to shape the polymer into a self-supporting membrane. Figure 7 lists some of the ways this is done with various polymers. The most commonly used procedure involves extrusion through a flat or circular die. If the polymer is thermoplastic, it can be heated to decrease the viscosity, or, if crystalline, to melt the material for extrusion into sheet form. Alternatively, the polymer may be "solubilized" as a viscous solution and extruded in that manner. Some films are made by extruding one polymer structure in sheet form and then post-treating the sheet chemically, or with heat to change the chemical composition. For example, this technique is used to make polyimide films<sup>4</sup>. Polymer films can also be polymerized onto a substrate from monomers in vapor form, or formed by the coalescence of polymer dispersions.

Superimposed upon the many ways that films can be formed are methods for combining polymers, adding particulates, chemical agents and post-extrusion operations. All can be considered under the topic of "processing for properties". Figure 8 lists just a few of the techniques that are used to achieve particular combinations of properties in films. To discuss each is beyond the scope of our discussion. Therefore, it is probably more informative to discuss one particular polymer system where mechanical and thermal processing methods are used to develop a wide variety of properties.

I have chosen to discuss the processing of poly(ethylene terephthalate) films. Many PET polyester films are manufactured

worldwide and sold under such names as Mylar®, "Melinex"<sup>1</sup>, "Celanar"<sup>2</sup>, "Lumirror"<sup>3</sup>. They serve many important markets for industrial and packaging applications.

Commercial PET is made by the transesterification of dimethyl terephthalate (DMT) with ethylene glycol or by the direct esterification of terephthalic acid with ethylene glycol. Three polymer processes are used to make the polymer; batch autoclave, continuous melt, and solid phase polymerization. The end result is a viscous polyester melt with the chemical structure shown in Figure 9. The average degree of polymerization is about 75 with a number average molecular weight of 14-15,000. Three aspects of the molecular are important, as shown in Figure 10:

- 1) It is a flat, planar molecule as shown on the model.
- 2) The carbonyl groups in the trans conformation form a linear chain without "kinks".
- 3) It can crystallize.

This is all that will be discussed about polymer synthesis, because the key concepts reside in what we can do with this polymer.

Figures 11-12 illustrate the commercial film-forming processes for PET<sup>5</sup>. These included melt extrusion of the polymer, quenching the melt to a "glassy" state, orientation and then thermal stabilization or relaxation.

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<sup>1</sup>Registered Trademark of ICI

<sup>2</sup>Registered Trademark of Celanese

<sup>3</sup>Registered Trademark of Toray



### Extrusion & Quenching

Molten polymer is extruded from a flat die. Since the viscosity of the molten polymer is of the order of 2500 poises at 280°C, a self-supporting film can be maintained over the space from the die to the cold quenching drum. Here, the film passes quickly from a fluid to a glass which has a density of about 1.33 g/cm<sup>3</sup> and the description of a cast, amorphous film is generally applied. The extruded polyester film, if cooled slowly, will crystallize spherulitically and become hazy, brittle and difficult to stretch. Consequently, the maximum attainable thickness of amorphous film depends on the rate of polymer crystallization and on the heat transfer rates attainable in quenching. Ultimately, the thermal conductivity and the crystallization rates become the limiting factors in the ability to produce a homogeneous glassy structure. This amorphous film can now be oriented by stretching or rolling.

The properties that we develop depend upon the way we utilize the viscoelastic properties of the glassy material. Temperature has a major role in the deformation processes of amorphous polyester films. At low temperatures molecular flexibility is suppressed, but at a sufficiently high temperature the material passes from a relatively rigid glass to a flexible, ductile viscous state. The temperature at which this transition occurs is commonly referred to as the glass transition temperature ( $T_g$ ). For PET the  $T_g$  is about 70°C and the film is generally processed at temperatures in the range 80 to 90°C. At these temperatures the film can be drawn or rolled easily. During the deformation of the heated film the

stresses developed cause the uncoiling and translational motion of the molecular chain segments. Concurrently, nucleation and strain-induced crystallization occur. Therefore, the net result of the deformation process is the partial alignment of molecular segments in the direction of deformation, coupled with the formation of a new phase.

Orientation by stretching may be accomplished in the direction of film travel (machine direction), at right angles to the direction of film travel (transverse direction), or in both directions. Stretching in the machine and transverse directions may be carried out simultaneously, for example, expanding a tubular film, or sequentially. Film can be stretched under conditions in which the product will shrink as much as 50% of the stretched dimensions on being heated above the processing temperature. This is sold as a heat-shrinkable polyester film. Film can also be heat set or crystallized while restrained and will exhibit much less residual shrinkage (ca. 1% at 105°C) on being heated. A dimensionally stabilized film with still less residual shrinkage (ca. 0.1%) can be prepared by heating the film under conditions of low stress (10 to 300 psi) to temperatures above the required-use temperature. Consequently, many process variations are used which produce different combinations of polyester film properties. For illustrative purposes we will discuss a sequential stretching process for making a biaxially stretched film having balanced physical properties.

### Stretching and Heat Setting

Amorphous film is preheated above the glass transition temperature and passed between two sets of heated nip rolls. Stretch or draw is developed by having the speed of the second set of nip rolls two to six times faster than the first set. Alternatively, stretching may be done over a series of heated idler rolls flanked on the entrance end by driven slow rolls and at the exit by driven fast rolls. The force required for stretching is a function of the film thickness, temperature and amount and rate of deformation.

Qualitatively, we can begin to understand what is happening during stretching by examining the stress-strain behavior of the polymer at the temperatures of interest. Figure 13 indicates three regions that occur. Region A up to the yield point of the stress-strain curves can be thought of as the work required to move the molecular segments from their equilibrium positions, that is, simple elastic extension. Region B is indicative of massive viscoelastic deformation of the film in which the molecular chain segments rotate, translate, and unfold and become aligned in the direction of draw. Once the system is set into motion little additional force is required. Ultimately, however, the ordering process is complete enough to require a greater force for further deformation and a Region C of reinforcement occurs. The system then becomes more ordered, increasing with the position on the ascending portion of the curve. The reinforcing portion of the curve looks like that observed in the strain-hardening of metals caused by the motion and aggregation of dislocations. Finally the stress will exceed the strength of the film and fracture occurs. The shape and

magnitude of the stress-strain curve depend on the temperatures used and on many other process parameters as well. Commercial deformation methods are designed to utilize these concepts to orient the structural elements in the film in one or more directions to produce the level of properties required.

Figure 14 represents a composite polyester film line. After the first direction stretch the uniaxially drawn and partially crystalline film then passes into a transverse direction stretcher called a tenterframe. This equipment is an adaptation of textile processing machinery and consists of film gripping devices attached to a moving endless chain. Film is transported by the chain through a series of independent zones which stretch and heat or cool the film. As the film leaves the longitudinal or machine direction stretcher (A), it is positioned automatically in the gripping devices and transported into a zone in which the film is again heated to increase the ductility. The chain path then diverges and the film is stretched in the transverse direction (B). Often the chain holding the film-gripping devices is attached to movable supports which permit variations to be made in the amount of transverse stretch. Typically the amount of transverse stretch is controlled to provide relatively balanced mechanical properties in the film. At the end of the stretch zone the doubly drawn film sheet exhibits high mechanical properties in both directions; it is tough and flexible but will shrink if heated above the stretching temperature. To decrease the tendency of the film to relax dimensionally it is crystallized under restraint in one or more zones by utilizing temperatures in the range of 150 to 230°C (C,D). This process, called heat setting,

increases the film density to about 1.39 g/cc. Typically, a heat set film may still shrink slightly (fractions of a percent) when heated above 80°C because of residual strains. This shrinkage can be reduced further by heating the film to a temperature lower than the heat set temperature but with little or no restraint (E). Before winding the film onto a roll it is cooled to ambient temperature and the film edges that were in the gripping devices are cut off (F).

A tensilized film (i.e., one more highly oriented in one direction than the other) can be made by additional processing of the biaxially oriented film. This is done by stretching again, usually in the machine direction.

### Film Morphology

It is worthwhile to digress for a moment to clarify the concept of orientation. In my experience, this term has generated vast amounts of confusion in communication when discussing polymer structure.

A convenient model to use when describing the crystalline texture of a polymer like PET is the fringed-micelle shown in Figure 15. In this model we visualize the film as composed of an imperfect two-phase system of interconnected crystalline and amorphous domains. The multiplicity of micro crystalline regions are tiny and are called crystallites. We say nothing about their internal structure except to recognize each as being composed of many unit cells. For the PET crystal (Figure 16) we would like to know how the direction of the chain axis and crystal axes change with stretching.

We would also like to know how properties depend on the way these structural entities are oriented in the film.

An elementary but useful approach was used to define orientation in films. The direction of stretch and the surface of the film may be used as a frame of reference that define a coordinate axis and a plane. Next we need to measure a structural parameter that describes the direction of molecular chain axes, crystal axes or crystal planes relative to the frame of reference chosen. In the limit, we can describe 6 major ways that these structural parameters are positioned in a film, as shown in Figure 17. Three of these (uniplanar, axial, uniplanar-axial) orientation types are important to the structure and properties of PET films. Orientation will not be perfect, and distributions of the directions of crystallite planes or axes will be observed. The definitions proposed are the limiting cases, designed to provide a simple and uniform terminology for describing orientation.

The structure of PET films has been studied with x-rays, infrared, nuclear magnetic resonance, optical diffraction, chemical and other classical techniques. One of the easiest methods used to follow the progress of orientation in transparent films is the refractive index. The only equipment needed is a suitable refractometer or a polarizing microscope with compensators. Amorphous PET film, stretched or rolled in one or both directions, becomes birefringent, thus indicating a preferred alignment of polarizable groups. A distribution of refractive indices exists which depends upon specific combinations of the process parameters used in making films. With

this background, we can now begin to describe structurally what occurs during the stretching and heat setting processes and relate these to the properties produced.

Measurements of the orientation by simple techniques such as the refractive index, along with measurements of the properties of stretched films reveal some unexpected characteristics. Figure 18 relates the average tensile strength to the amount of stretch while Figure 19 relates the % elongation to the total molecular orientation as inferred from the refractive index in the 3 orthogonal directions. Something of significance has occurred at a deformation of about 250% (3.5 x deformation ratio).

Use of more definitive methods (x-ray & infrared) to measure the structure provides further insight into what happens to the film. Stretching PET in one direction is found to produce axial-uniplanar orientation i.e., the chain axes or "C" axes of the crystals tend to become aligned in the direction of stretch and concurrently the aromatic rings and (100) crystal planes tend to become parallel with the film surface. The parallelism begins to change very rapidly at about 250% deformation under the process conditions used. In the limit, we obtain a structure that is somewhat like a deck of cards. In a fiber, we would expect to find a structure not unlike the growth rings in a tree.

Stretching in the second direction redistributes the structure toward the new direction of stress. Ultimately we arrive at a balanced structure in which the aromatic rings and the (100) crystal planes

are mostly parallel to the film surface but the chain axes or "C" axes of the crystals are distributed more or less uniformly between the two orthogonal stretch directions. In the limit, we now have developed a uniplanar structure. The uniformity and the level of properties generated will depend upon how much we have stretched, the temperature, the strain rate and geometrical factors of the equipment used.

The film we have made will be heat shrinkable and unstable dimensionally unless we heat-set it. The amorphous polymer we extruded and quenched had essentially 0% crystallinity. When we stretched it in the first direction we developed strain induced crystallinity which we measure by flotation density methods. Again, assuming our 2-phase fringed-micelle model we find the one way stretched film has about 20% crystallinity depending upon how we stretched it. During the second direction stretch we have developed about 5% more crystallinity. Heat setting is a process by which the film is restrained and exposed to temperatures in excess of the maximum use temperature. A complex process of crystallite melting, crystallite growth and structural rearrangement occurs which tends to stabilize the structure. During heat setting the crystallinity increases by another 20% and the result is a film with decreased shrinkage and about 45-50% crystallinity. It is highly lamellar as shown in Figure 20.

If a greater level of dimensional stability is required, the heat set film can be thermally relaxed. Since the film is constrained during heat setting, residual stresses remain that can be relieved by heating the film unrestrained at a lower temperature



than that used for heat setting. A new equilibrium structure is produced having improved dimensional stability.

Many variations of these stretching and thermal processes have been developed to tailor-make properties for specific uses. One way of illustrating the breadth of mechanical properties that can be produced by this one polymer system is by examination of the variety of force-elongation curves. These are the curves obtained on a tensile testing machine by pulling the films in orthogonal directions (Figure 21). The magnitude of the tensile properties that are achieved is shown in Figure 22.

#### Surface Properties

Most polymer films are given post-treatments of various kinds, either by the film manufacturer or by a converter. Among these treatments are coatings for protection against water and gases, magnetic oxides for recording media and metals or polymer systems for specific uses. Figure 23 is a partial list of the surface properties that are important to films either singly or in unique combinations with others. Some are mutually exclusive and "processing for surface properties" involves choosing the best compromise.

Throughout manufacture, conversion and use, the film must possess a property called "handling" which is difficult to define precisely. Good "handling" infers that the film can be wound at high speeds without damage to produce an attractive product, or be shoved off a pile of sheets without jamming processing machinery.

Many parameters influence "handling". Among the most important are thickness uniformity (across and along the sheet), surface topography, hydrodynamic and boundary friction, and the stiffness of the web. Since rigidity is proportional to the cube of the thickness, the nominal thickness to be wound and the mechanics of winding machinery also influence roll aesthetics.

Plastic films with good optics are prized for their smooth, glossy surfaces. It is easy to damage the surface and yet it is essential that films be wound into large rolls at high speeds and have a pleasing appearance. In the jargon of the film business, the properties called "slip" and "blocking" are important to the way films can be used. "Slip" is an imprecise term that infers the ability of films to slide over themselves. As a roll winds, it is necessary for each layer to tighten and to cinch upon itself. It must stay in registry with the layer underneath, and also accomodate large perturbations in winding speeds, vibrations and tension without forming wrinkles or damaging the surfaces. Alternatively, the film cannot be so slippery that the roll telescopes. A thick, stiff film presents one set of winding problems, and very thin films, i.e. less than 1/1000 inch thick, other types of problems. "Blocking" is the tendency of films to stick together. Often the surfaces are so smooth that they tend to fuse together and part only when the surfaces tear. From these brief descriptions of slip and blocking, it becomes obvious that surface topography and friction receive much attention in the film business.

## Friction

Early work by the Frenchman, Amontons, suggested that frictional effects in metals were a consequence of surface roughness. It was believed that the points of roughness or asperities interlocked and the force of friction was that necessary to overcome the interlocking. Others held the opinion that friction was a consequence of surface adhesion. Still others have held the opinion that frictional forces are a consequence of one surface ploughing through the other. Since most of the knowledge of polymer friction is an extension of the theories of metallic friction, we should contrast and compare these, and seek out the similarities and differences.

Metallic friction can be explained in terms of adhesion which embodies the principles given in Figure 24. The expression  $\mu = s/p$  states that the coefficient of metallic friction is determined solely by material constants independent of load, size or surface finish. Two types of friction are important, static and kinetic. The static coefficient of friction ( $\mu_s$ ) represents the ratio of the peak force to load required to initiate sliding, while the kinetic coefficient ( $\mu_k$ ) represents a similar ratio required to produce continuous sliding. Metals normally show little difference between  $\mu_s$  and  $\mu_k$ , but many polymers exhibit much higher static coefficients. Moreover,  $\mu_s$  for polymers depends upon the time that the surfaces have been in contact.

Many workers have shown several important differences between the frictional effects of metals and polymers even though the adhesion theory appears to be useful in both systems. Figure 25

compares the frictional effects. Amontons' law  $F=\mu W$ , is modified to account for the visco-elasticity of polymer asperities. With polymers the area of real contact that occurs is proportional to  $W^n$  where  $n=1$  for complete plastic deformation and  $n=2/3$  for elastic deformation. For many polymer systems ( $n$ ) is intermediate between these values and is influenced strongly by the deformation mechanism. The coefficients of static friction of polymer films (film surfaces against each other), range from values greater than 1.0 to that of Teflon® at about 0.04. It is common practice in film technology to change the measured  $\mu_s$  by altering the area of contact. This is done by the techniques shown in Figure 26. Polymer additives that increase surface roughness and produce asperities ranging in height from 1-100 microinches are titania, silica and clays of various kinds. Typical changes that occur in the surface roughness are illustrated in Figure 27. Alternatively, sizings such as chalk, clay, starch are often used to keep the surfaces apart. All of these ultimately tend to destroy the optics of the film and a compromise always results. Particle size, concentrations, toxicity and the economics for polymer recovery often dictate the technology to be used.

We can also lubricate the surface. Often, a slip agent can be added to the polymer melt, which exudes to the surface of the quenched film<sup>6</sup>. This technique has had great success with some polyolefins where the amides of fatty acids are often used. Many proprietary formulations of slip, antiblock, antistat and anti-oxidant formulations have been developed and the patent and technical literature is replete with examples.

With a lubricant between the surfaces, the total weight is supported by 2 kinds of junctions:

- High shear junctions between the original surfaces.
- Low shear junctions between the original surface and the lubricant and also between the lubricant-lubricant junctions.

$$F = A[\alpha S_C + (1-\alpha)S_1]$$

$\alpha$  = fraction of (A) not covered by the lubricant

$S_C$  = shear strength of base material junctions

$S_1$  = shear strength of junctions between lubricated surfaces

Fort<sup>7</sup> has enumerated some general guidelines for the boundary lubrication of PET. These guidelines are also pertinent for other polymers:

- a) Desirable to have a lubricant with very low shear strength.
- b) High surface coverage helps.
- c) Solid lubricants are more effective than liquid lubricants.
- d) Non-bonded lubricants are transient.
- e) Friction increases with temperature and increases substantially above the melting point of the solid lubricant. Very high melting solid lubricants are desirable.
- f) Abrasion and wear become more severe as the static coefficient of friction ( $\bar{\mu}_s$ ) increases.

If optical properties are relatively unimportant, the surface area of real contact can be changed by embossing the film, roughening the surface with a brush, sandblasting the surface with an abrasive and by chemical etchants. At the other end of the frictional "spectrum" are the smooth, very flat surfaces that tend to adhere together and cause "blocking". At every point of contact interfacial damage occurs. As a last resort, interleaving with an innocuous layer such as glassine paper can be utilized to separate the surfaces.

Still another variable is significant to our understanding of winding. We wind in air, which becomes trapped between the layers of film and acts like a lubricant. If a large amount of air is included, film-to-film contact becomes negligible; the film rides on an air bearing and the winding system becomes either essentially uncontrollable or the air layer will limit production speeds<sup>8</sup>. Film surface roughness, winding tensions, lay-on rolls and many other methods are utilized to control these air bearing effects. The technology of winding is a science in itself.

The term "keying" has been used to describe the chemical character of the polymer surface and its ability to accept printing inks and other coatings. Many polymer films have low surface free energies which are often incompatible with the wetting of other materials that need to be applied onto them.

It is generally agreed in theory that secondary and hydrogen bonding forces are sufficient to produce good adhesion without the

need for primary chemical bonding. These secondary valence forces decrease as the inverse  $1/6$  power of the distance between molecules. Therefore, surfaces to adhere well should come into good wetting contact.

Zisman and co-workers have developed an empirical parameter called the critical surface energy  $(\gamma_c)^9$ , which helps the prediction of what material is likely to wet another. Liquids with surface energies greater than  $\gamma_c$  are not likely to spread over the solid. Typical critical surface energies for a number of polymers are shown in Figure 28.

It is difficult to wet polymer solids with water and two general techniques are used to aid interfacial contact. Often the film is primed by making it polar (higher  $\gamma_c$ ) by means of an electrical discharge or by flame treatment. Alternatively, wetting agents are added to the coating compositions which decrease the surface energy to a value equal to or below that of the polymer film. The unquestioned commercial successes of coating technology to "process for properties" have enriched our way of life.

The kinds and numbers of coatings, adhesives and other surface treatments used on films are what make them important to us. Reflect for a moment on the combinations that are possible from polymer and metallic coatings, and from the coextrusion systems and laminate technology now in use. A wealth of possibilities exist for the development of polymer films for packaging and for engineering materials.

Finally, what can we expect in the future?

Just a brief survey of the technical literature indicates substantial growth in the ability and sophistication to manipulate polymer properties. Concurrent with advances in the understanding of polymer morphology, we are witnessing the development of tougher materials formed by the blending of inexpensive polymers. High strength to weight ratios and excellent fatigue resistance are likely goals. Processing technology will also be an important part of the future of films. Advanced techniques of coextrusion, multiple coating methods and improved ways to control orientation and crystallinity are certain to be major contributors to the films of the next decade.

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Attachments



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FIGURE 1

FUNCTIONS OF POLYMERIC FILMS

Enclosure

Cover and protect  
Barrier to gases or perm-selective

Carrier

Information storage & retrieval  
Light, sound, visual symbols  
Coatings of various kinds  
    abrasive  
    optical  
    adhesives

Electrical Component

Insulator  
Dielectric

Structural Member

Composites

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FIGURE 2

PROPERTIES IMPORTANT TO MICROFILM BASE

Bulk Properties

Stiffness - thicknesses 3-7 mils  
Optical Clarity and Uniformity  
Accept Coatings Uniformly  
Dimensionally Stable  
Free from Volume Defects

Surface Properties

Good "Handling"  
Thickness Uniformity  
Free from Surface Defects

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FIGURE 3

PROPERTIES IMPORTANT TO CAPACITOR TISSUE

Bulk Properties

Dielectric Constant  
Dielectric Breakdown  
Dielectric Loss  
  
Resistance -  
    Volume  
    Surface  
Thicknesses - 0.1-0.2 mils  
Mechanical Strength & Integrity (no pinholes)  
Dim. Stability

Surface Properties

"Handling" & Winding  
"Impregnation"

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FIGURE 4

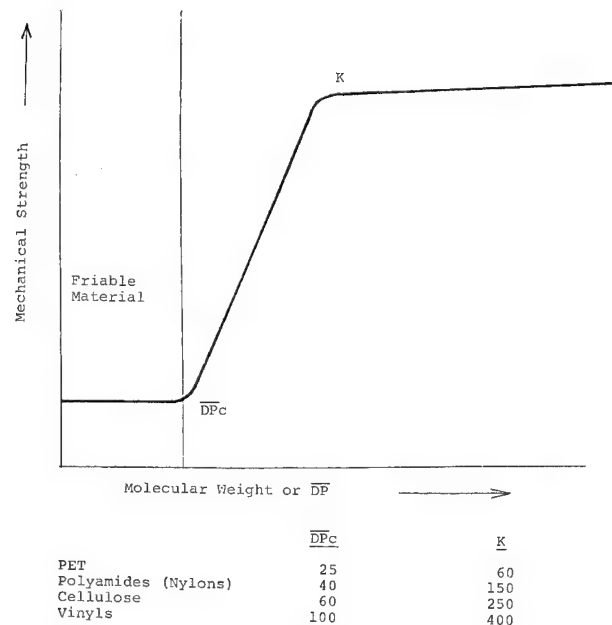


FIGURE 5

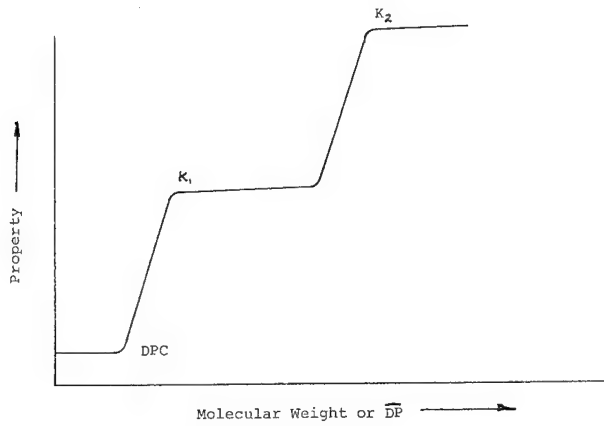
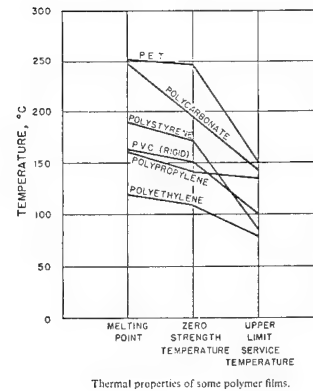


FIGURE 6



Thermal properties of some polymer films.

FIGURE 7

METHODS TO PRODUCE POLYMERIC FILMS

Thermoplastics -

- Melt Extrusion through a Flat or Circular Die (PET, nylons, PE, Ionomers)
- Batch
- Continuous

Solvent Extrusion through a Flat Die (CA, Cellulose Xanthate, PVF)

In Situ Polymerization (PMMA, epoxies)

Calender (PVC, polyurethanes)

Polymer 1 (extrusion) + Post Reaction to Polymer 2 (polyimides)

Vapor Deposition - (polyxylylene)

Casting onto a Substrate - { Electrodeposition  
Dispersions

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FIGURE 8

PROCESSING FOR PROPERTIES

OPTIONS

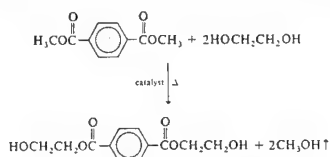
Polymer Systems

- 1) Copolymerization
- 2) Coextrusion
- 3) Additives
  - Lubricants
  - Flame Retardants
  - Nucleating Agents
  - Fillers
  - Stabilizers
  - Anti-oxidants
  - Ultraviolet absorbers
- 4) Mechanical and thermal processing
- 5) Post-processing
  - Coating
  - Radiation
  - Chemical Post Reactions

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FIGURE 9

1. Transesterification of dimethyl terephthalate with ethylene glycol.



2. Direct esterification of terephthalic acid and ethylene glycol.

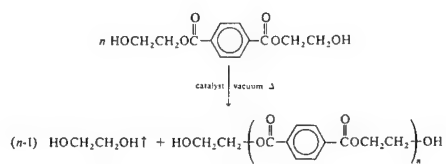
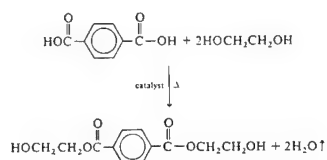


FIGURE 11

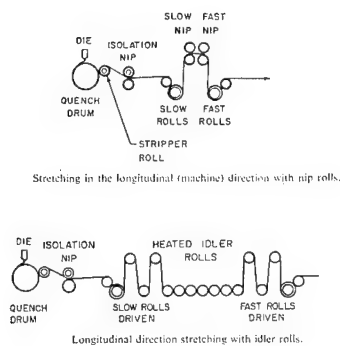


FIGURE 10

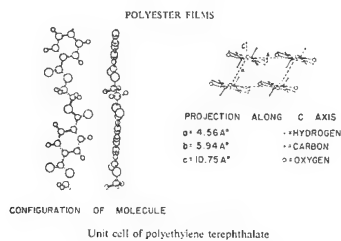


FIGURE 12 & 14

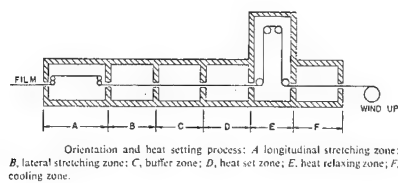


FIGURE 13

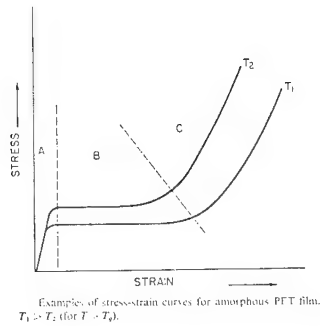


FIGURE 17

NAME	GEOMETRY	REFERENCE	OPERATOR	SCHEMATIC STRUCTURE	POLE FIGURE
RANDOM	NO ORIENTATION	NONE	NONE		
PLANEAR	GIVEN CRYSTAL AXIS PARALLEL TO A REFERENCE PLANE	PLANE	AXIS		
QUADRANT	GIVEN CRYSTAL PLANE PARALLEL TO A REFERENCE PLANE	PLANE	PLANE		
AXIAL	GIVEN CRYSTAL AXIS PARALLEL TO A REFERENCE AXIS	AXIS	AXIS		
PLAN-AXIAL	GIVEN CRYSTAL PLANE PARALLEL TO A REFERENCE AXIS	AXIS	PLANE		
UNIPOLAR-AXIAL	GIVEN CRYSTAL AXIS PARALLEL TO A REFERENCE AXIS AND A GIVEN CRYSTAL PLANE PARALLEL TO A REFERENCE PLANE	AXIS	PLANE		

Classification of orientation types

FIGURE 15

FRINGED - MICELLE MODEL OF STRUCTURE

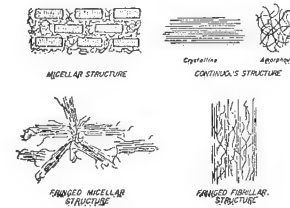


FIGURE 16

PET CRYSTAL UNIT CELL

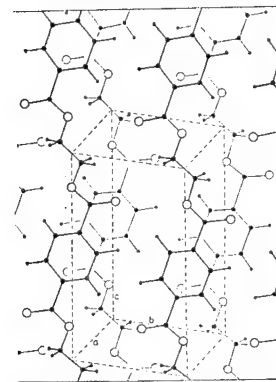


FIGURE 18

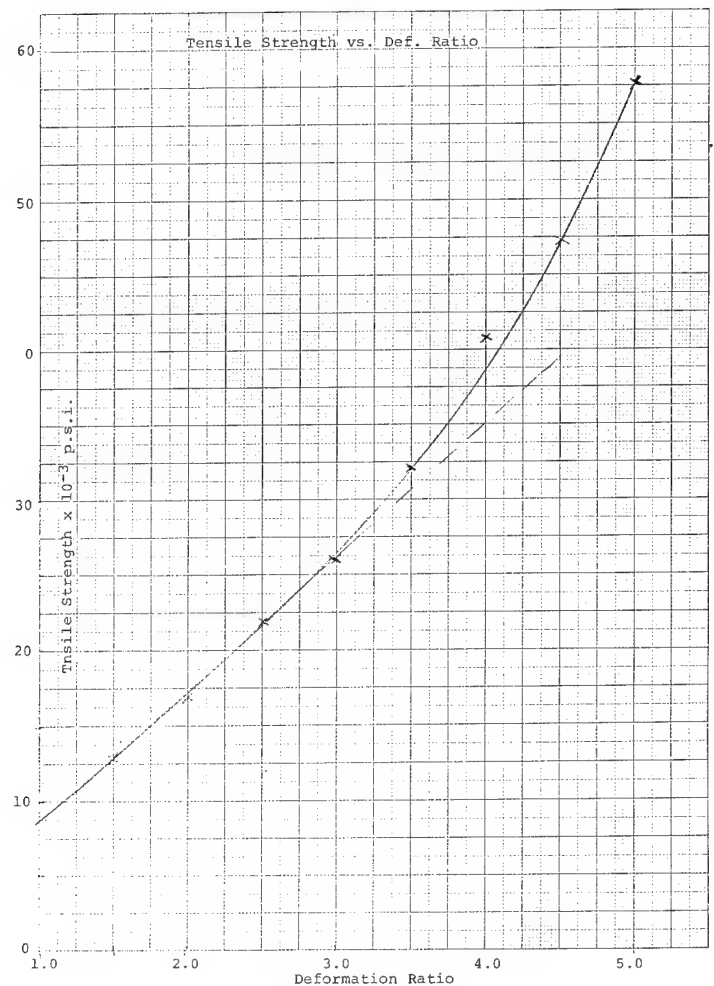


FIGURE 19

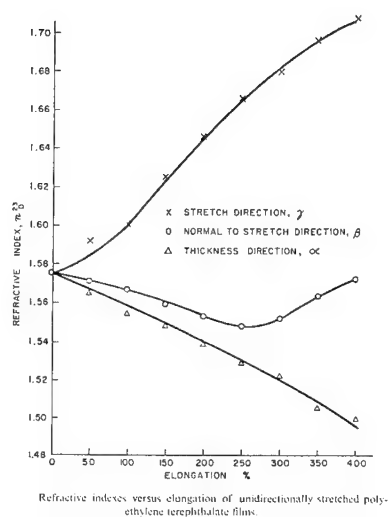
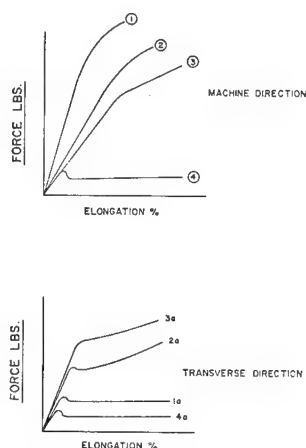


FIGURE 20



Lamellar Structure of  
Oriented Pet Film

FIGURE 21



Force-elongation curves of polyethylene terephthalate films having various levels of orientation and crystallinity: 1, uniaxially stretched only; 2, tensile film; 3, biaxially stretched; 4, "amorphous" unoriented.

FIGURE 22

PROPERTIES OF VARIOUS PET FILM STRUCTURES

	Amorphous	1wx	2wx	2wx ps
Modulus, psi	350,000	1,270,000	650,000	1,000,000
	350,000	253,000	650,000	500,000
Tensile Str., psi	7,500	41,000	25,000	38,000
	7,500	7,000	25,000	17,000
Elongation, %	>500	48	120	52
	>500	445	120	250
Force @ 5% Elongation	-	33,000	14,500	26,500
	-	7,500	14,500	11,000

1wx = One way stretched

2wx = Two way stretched

2wx ps = Two way stretched + post stretch

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FIGURE 23

SURFACE PROPERTIES

- A) "HANDLING"
- Coeff. of hydrodynamic and boundary friction
  - Surface topography
  - Lubricity
  - Tracking
  - Uniformity - gauge
  - Stiffness
- B) Optical
- Haze
  - Clarity
  - Gloss
- C) Energy
- Wetting
  - Adherability
  - Sealability
  - Resistivity & conductivity
  - Barrier integrity
- D) Toxicity
- Chemical constitution
  - Chemical stability

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FIGURE 24

PRINCIPLES OF METALLIC FRICTION

- All surfaces are rough.
- Contact area is governed by asperity deformation.
- Junction formation occurs at the contact points.
- Friction is the shearing force necessary to break the junctions.

Area of Contact = Weight/yield pressure of asperities

$$A = W/p$$

Force = (Area) (Shear Strength)

$$F = As = \frac{(W)}{P}(S)$$

$$F = \mu W$$

$$\mu = \frac{F}{W} = \frac{S}{P} \quad \text{a function of material properties}$$

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FIGURE 25

FRICTION EFFECTS

Metals

Independent of load

Independent of roughness

$$F = \mu W$$

Polymers

Increases as load reduced

Decreases with surface roughness

$$F = \mu W^n$$

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FIGURE 26

Methods to Change the Frictional Characteristics of Polymer Films

- Reduced Area of Contact
  - Additives that produce asperities
  - Surface Abrasion
    - Brushing, Sandblasting
    - Embossing
    - Chemical Etchants
- Surface Lubrication
  - Additives that migrate to the surface
  - Sizings
    - Organic, Inorganic
    - Bonded Lubricants
- Coat on a New Surface

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FIGURE 27

SURFACE ROUGHNESS  
OBTAINED FROM POLYMER ADDITIVES

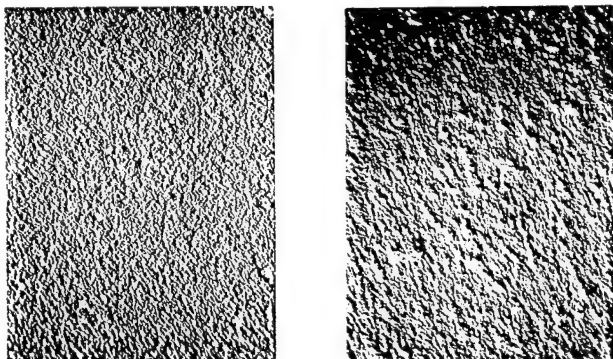


FIGURE 28

SURFACE ENERGY OF FILMS

	<u>Water Contact Angle</u>	<u><math>\gamma_c</math> dynes/cm</u>
Nylon 6-6	72	46
Polystyrene	91	43
Poly(ethylene terephthalate)	76	43
Poly(vinylidene chloride)	80	40
Poly(vinyl chloride)	87	39
Poly(methyl methacrylate)	80	39
Polyethylene (low density)	104	31
Paraffin	108	23
Poly(tetrafluoro ethylene)	108	18.5

Surface Energies  $\gamma$  dynes/cm

Water - 72.8

Methylene Iodide - 50.8

Carbon Tetrachloride - 26.8

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## Man-Made Fiber Processing: Relation to Properties

James P. Bell

### Synopsis

A discussion of the synthetic fiber processing variables of orientation, shear crystallization, and crystallization temperature is presented, with emphasis on the effects of these variables on polymer morphology and resulting tensile properties. The discussion is slanted to provide new ideas as to how these variables can be beneficially used in plastics processing.

Since most synthetic textile fibers are based on polymers which are also used for molding and extrusion applications, fiber technology and plastics technology are related. Typical synthetic fibers are based on nylon 6 and 6,6, polyesters, and poly(acrylonitrile). One can gain new ideas for improvement of fiber properties from research on plastics, and vice versa.

This paper deals with three especially important parameters in fiber processing, and the effects of these parameters on selected properties. From these effects we can hopefully make inferences concerning processing of thermoplastics. The variables are molecular orientation, shear crystallization, and crystallization temperature.

#### Molecular Orientation

Orientation of the polymer chains parallel to the direction of the applied stress can result in as much as a ten-fold strength improvement, as seen from figure 1.<sup>(1,2)</sup> One gram/denier in nylon 6,6 is equivalent to approximately 14,500 psi\*, and in poly(ethylene terephthalate) approximately 17,400 lbs<sub>f</sub>/in<sup>2</sup>. Steel has a strength of approximately 65,000 lbs<sub>f</sub>/in<sup>2</sup>. If one considers a drawn textile yarn with an average tenacity of 5 gms/denier, and dividing each strength by the specific gravity so that strengths are expressed on an equal weight of material basis:

Nylon	63,600 psi
PET	63,000 psi
Mild annealed steel	8,242 psi

---

\*( $\sigma$ , psi) =  $(9 \times 10^5)$  (density, gms/cc) (applied stress, gms/denier)

On an equal volume of material basis:

tenacity 5 gm/den. nylon 6,6	72,500 psi
" " PET	87,000 psi
mild annealed steel	65,000 psi

Clearly, the axial property improvements obtained by orienting the molecules in fibers are a vital factor in their success, and one begins to think of means to obtain controlled orientation in plastic parts and articles. One observes from figure 1 that considerable differences in tensile properties are obtainable for the same chemical structure. (See, for example, the different curves for nylon 6,6.)

Molecular orientation in fibers is usually obtained by stretching the fiber at a temperature in the general range of the undrawn polymer glass transition temperature. If a much higher temperature is used initially, the molecules have time to relax and disorient before they can be "frozen" in their oriented state. If too low stretching temperature is used, the fiber behavior will be too brittle and rupture will occur.

Tire cord products are usually made by a two-stage draw. In the first stage the molecules are partially crystallized in the oriented state. The crystals, somewhat like cross-linkages, prevent randomization when the non-crystalline linkages are further oriented by stretching in a second operation at a higher temperature. The symbol HT on Fig. 1 designates these "high tenacity" products.

The oriented fiber will of course be no stronger than its weakest point. A highly oriented, strong region is of little value if there is a weak region next to it supporting the same stress, since failure will be determined by the weak region. The same philosophy applies to a plastic part; it can only be as strong as its weakest link. Uniformity of orientation is therefore very important.

To obtain the dramatic improvement in axial strength and modulus we must accept a decrease in properties perpendicular to the direction of orientation. Examples of this are shown in Fig. 2<sup>(3-5)</sup>, in which the modulus perpendicular to stretch decreases as the draw ratio increases. The draw ratio is the ratio of the final stretched length to the initial length. The intercrystalline molecules are tightly stretched in the axial direction, somewhat like stretched, frozen rubber bands. As one might expect, the fiber becomes more sensitive to nicks, scratches and abrasion. In the fiber industry there are many examples of special heat treatments, etc., designed to partially restore lost transverse properties.

Another consequence of orientation is shrinkage, Fig. 3<sup>(6)</sup>. When the molecules lying between crystals are heated above their glass transition they will attempt to retract to a random, higher entropy configuration. This is the reason for shrinkage of garments of synthetics that have not been given special stabilizing treatments, and it is also the reason for distortion of many amorphous plastic articles heated above their glass transition temperature. It can also contribute to distortion during fabrication of plastic articles.

Nylon and polyester oriented fibers have substructures like rope, called macrofibrils. The macrofibrils contain microfibrils, which in turn contain the crystalline lamellae and individual chains. Fig. 4<sup>(7)</sup> shows a drawn polyester fiber that has been nicked with a razorblade, and the surface layer pulled back with microtweezers. The work is done under a microscope, since the fibers are only 20 microns, smaller than a fine hair, in diameter. The macrofibrils are visible bridging the gap between the core and the surface pulled away, as seen on Figs. 5 and 6. The fiber in Fig. 6 was peeled at low temperature, which accounts for its more brittle appearance. Fig. 7 is a scanning microscope photograph of a fiber end after fracture<sup>(7,8)</sup>. The macrofibrils again appear visible.

In the overall view, although shrinkage and transverse property loss are problems, the axial property improvements from orientation are great enough to be useful, even vital for synthetic fibers.

During fiber spinning the molecules are oriented parallel to the direction of flow in the spinnaret or capillary, but melt swelling occurs at the orifice exit and the orientation is almost completely lost. The orientation ~~was~~ found in "spun" yarn, i.e., yarn from the spinning process, is almost entirely the result of stretching of the melt between the "die swell" region and the solidification point. For this reason spun yarn orientation can be controlled to a considerable extent by controlling the ratio of the linear velocity at spun yarn windup to the linear velocity at the spinnaret, which changes the tension or "stretching force" on the melt. This is illustrated in Fig. 8<sup>(9)</sup>. Generally speaking, low spun yarn orientation is desired; orientation is more effectively introduced in the subsequent cold drawing process at a temperature near  $T_g$ .

From the foregoing considerations one might rationalize that axial orientation in extruded tubing could be obtained by stretching the melt before solidification, but that this axial orientation would be accompanied by a loss in burst (circumferential) strength. Shear with cooling is necessary. In filling an empty cavity, shear at the melt-gas interface is usually low and orientation is lost. Flow lines near gate and other flow-through areas are indicative of orientation, but this may be only near the mold surfaces; the material in the center of the cross-section may be at a higher temperature and thus it becomes randomized more easily. Gating to take advantage of flow-through orientation, and also flow-through designs in which the melt flows completely through the part, have merit where orientation is desired.

## 2. Shear Crystallization

Cooling of a crystallizable, quiescent melt results in an entirely different morphology than cooling of the same melt in a shear field. Shearing enhances the probability of alignment of several chains to form a nucleus of critical size, which in turn results in the formation of very many nuclei. Without shear, nucleation is much slower and occurs most frequently at foreign surfaces, such as at trace impurities or dirt. The net effect of this difference in nucleation is that a fiber usually contains a large number of tiny crystallites interconnected by a large number of tie molecules, whereas in an injection molded part a relatively small number of nuclei grow into large spherulites.

The problem with this latter situation is that there are relatively few tie molecules extending between the large spherulites, and the inter-spherulite area is weak, somewhat like grain boundaries in metals. Fig. 9<sup>(10)</sup> shows clearly the boundaries between spherulites. Reding and Brown<sup>(11)</sup> and also Reinshagen and Dunlap<sup>(11)</sup> found that in poly(chlorotrifluorethylene) and polypropylene, respectively, cracks tend to propagate along the radii (between the radiating arms) of spherulites and also between spherulites. Large spherulites can also act as points of stress concentration, resulting in initiation of failure. Many tiny crystallites usually result in a stronger, more stable system. One may increase the number of nuclei in a melt by adding appropriate solid impurities as nucleating agents, but we really desire nuclei in the tens of angstroms size range, whereas most powders are much larger. If the number of particles is to correspond approximately to the number of crystallites we wish, then the concentration of the additive would be high enough to affect other properties and also probably the cost. Nucleating agents can help, particularly with melts that are difficult to crystallize, but it would appear that such a route is unlikely to result in the many tiny crystallites found after shear crystallization.

Because of the very many nuclei, the rate of crystallization during shear is up to 1000 times faster than for an undisturbed melt. This is illustrated on Fig. 10, for isotactic polypropylene<sup>(9)</sup>. Similar data are also available for high density polyethylene<sup>(9)</sup>. The symbol  $\sigma$  represents the shear stress in dynes/cm<sup>2</sup> for the two sets of data from melt spinning. Poly-(ethylene terephthalate), PET, crystallizes slowly and the crystallinity of normal PET spun yarn is less than one percent. Nakamura and coworkers<sup>(12)</sup> have found, however, that if the take-up velocity is increased markedly, resulting in stress, PET will crystallize. This can apparently be attributed to the increased crystallization rate associated with the melt strain. The same effect is not readily obtained by increasing the shear in the spinnaret, probably because much of the orientation is lost and nuclei inhibited from forming in the melt expansion or "die swell" region at the spinnaret exit.

One might comment that since the stress-to-break for unoriented nylon 6,6 fibers is of the order of one gm/denier, and this converts to:

$$\begin{aligned}\sigma_B &= (9 \times 10^5) \text{ (density) (Stress-to-break, gm/den)} \\ &= (9 \times 10^5) (1.14) (1) \\ &= 1.026 \times 10^6 \text{ gm/cm}^2\end{aligned}$$

$$\text{or } \sigma_B = 14,560 \text{ psi}$$

This  $\sigma_B$  is not much different from nylon plastic. In a way we are comparing apples and oranges, however, since the stresses are based on the original cross sectional area. The fiber has stretched perhaps 300% and the plastic perhaps 30%; the stresses at failure are vastly different when based on the actual cross section. Of course orientation has occurred in the fiber also, which makes comparison of effects due to the two morphologies alone difficult, if not impossible.

As with controlled orientation, control of the number and size of crystals by means of shear should offer improved, more uniform properties. Unfortunately, the difficulty of maintaining desired shear gradients is great, although perhaps feasible for some symmetrical shapes. If one examines the cross section of an injection molded tensile bar under polarized light, one commonly observes smaller spherulites near the surface where the shear gradient is higher and the temperature lower, and one finds large spherulites in the center where the shear profile is flat and the temperature remains high for a longer time. This center region is usually less resistant to weathering, boiling water, etc.

#### Crystallization Temperature and Uniformity

Temperature control during crystallization is vital in the production of crystallizable synthetic fibers. In melt spinning processes<sup>(13)</sup> not only is the melt temperature tightly controlled, but also conditions in the fiber formation region immediately following the spinnaret are held within narrow limits. A device called a chimney is used, which admits controlled temperature air into the cooling zone. Each manufacturer has his own design for the chimney region. The designs range from a closed box with simple counter-current air flow to a porous box or cylinder into which a controlled gradient of air is introduced through the walls. Rate of air flow, distribution of air flow, and air temperature are controlled to obtain desired spun yarn properties. Minor changes in air flow rate or distribution sometimes cause marked variations in the dye diffusion rate into the fibers, causing streaks in a fabric made from them. Other properties are also affected. If the outside of a fiber reaches its crystallization temperature substantially before the inside, the outside forms a solid "skin" while the inner core is still molten. An example of "skin-core" morphology in viscose rayon is shown in figure 11<sup>(14a)</sup>. The skin has been selectively stained. Similar effects are observed for nylons<sup>(14b)</sup>.



Since the "skin" and the "core" were crystallized under different conditions, they have different properties. "Skin-core" effects are also found in many plastic articles. The skin may be desirable or undesirable, but it is important to recognize that it is present and that the morphology is subject to at least some control.

An illustration of the effect of cooling rate on PET, which crystallizes slowly, is shown in figure 13<sup>(15)</sup>. The various curves show different rates of heating in the Differential Scanning Calorimeter, DSC. The PET is crystallizing during heating, before the melting temperature is reached. The rate of heating (or, correspondingly, cooling) is low, the crystallinity developed is greater. Greater crystallinity is shown by the greater area under the DSC exotherm in the melting range. In melt spinning of this particular polymer one wishes to avoid crystallization, so that cooling must be sufficiently rapid. Uniformity is still important, since local variations in temperature along the forming fiber will lead to local variations in orientation and thus other properties.

Cooling at different rates can also lead to DSC curves which exhibit two melting endotherms by DSC, particularly for nylon 6. These two endotherms vary in relative magnitude, depending upon the cooling rate. The nature of this two-melting-peak phenomenon has been discussed elsewhere<sup>(16)</sup>. Differences in cooling rates of PET spun yarn samples, which are essentially amorphous, have also been reported to have an effect on  $T_g$ , the glass transition temperature<sup>(15)</sup>.

The rate of crystal nucleation is a function not only of impurity concentration and shear, variables which were discussed in the preceding section, but also of temperature. As the temperature is lowered, the rate of nucleation increases because of the greater free energy driving force. One might guess

that the lower the temperature, the higher the crystallization rate. This is not true, however, since as the temperature is lowered the melt viscosity increases, and the growth process is inhibited. One actually observes a maximum crystallization rate at about 80% of the melting temperature<sup>(17)</sup>; this is a balance between increased nucleation and decreased growth as the temperature is lowered. Regions of a part which have cooled slowly, in the absence of shear, will tend to have few nuclei but rapid growth, resulting in large spherulites with an overall relatively high crystallinity. Thin regions such as fibers which have cooled rapidly will tend to have many small crystallites, lower overall crystallinity, but perhaps a stronger structure because of the greater number of connecting molecules. Reinshagen and Dunlap<sup>(11)</sup> have shown that the degree of undercooling and the number of intercrystalline tie molecules determine the physical properties, rather than the size of spherulites per se. Of course, when spherulite size is controlled by degree of undercooling, large spherulites mean few tie molecules. The mechanical and chemical resistance properties of a part which has sections undergoing different thermal histories can vary from section to section. It is possible to select the sections of critical importance and control the cooling rate in those sections, allowing also for shear effects.

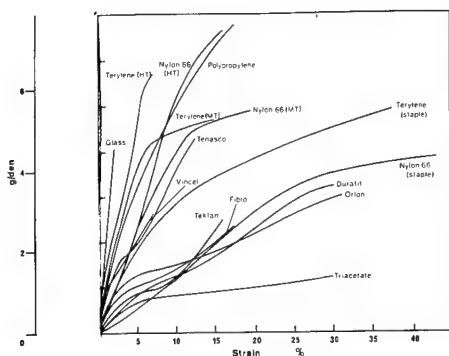
Heat setting is a process in which synthetic textile fibers are exposed to a temperature somewhat higher than they are expected to see in use. Exposure to the high temperature permits the molecules to wiggle and form inter and intra molecular bonds which tend to lock or "set" the structure; no dimensional changes should take place when the sample is subsequently heated to a lower temperature. The heat setting may be at fixed length, permitting no relaxation, or free, permitting some shrinkage and more crystallization to occur. Heat setting free is analogous to annealing of plastic parts in an unrestrained environment.

While the overall crystallinity increases on annealing, other changes can also take place, as shown by figure 13<sup>(18)</sup>. The PET yarns represented in this figure were annealed unrestrained for six hours at the specified temperature. The diffusion coefficient of a typical non-ionic dyestuff goes through a minimum at an annealing temperature of about 175°C. The explanation given<sup>(18)</sup> is that below 175° additional crystallization further restricts motions of tie molecules, making diffusion of the dye more difficult. Above 175° X-ray diffraction measurements of crystallite sizes show that the large crystallites are growing and the small ones are disappearing. The net result at these high temperatures is a smaller number of larger crystallites, and less restraint on the tie molecules connecting them. The lowered restraint enhances diffusion. The point to be made here is that crystallinity alone is not sufficient to define the morphology or associated properties. Just as the diffusion constant does not show a monotonic change with increasing annealing temperature, there are many other properties which will show similar behavior.

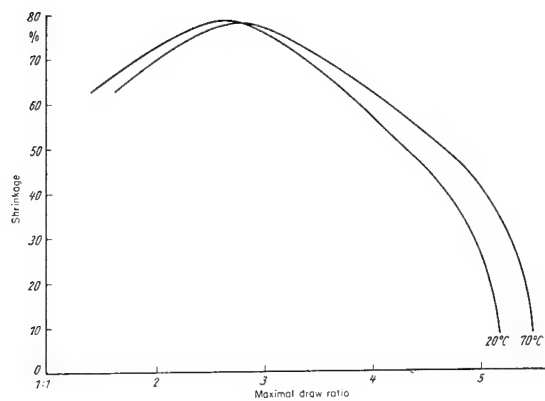
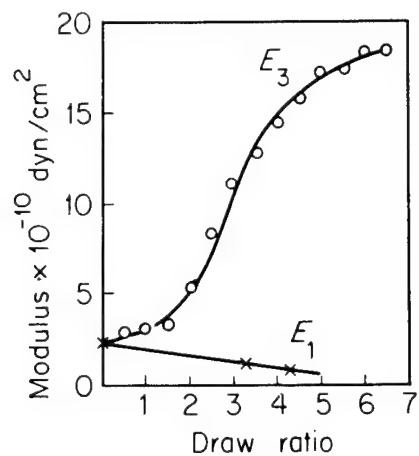
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Note: Viscose rayon variants are Fibro (regular-staple); Vincel (high-wet-modulus), and Tenasco (high-tenacity, industrial). Teklan is modacrylic; Terylene is polyester fibre; Orion is acrylic fibre



Boiling shrinkage of undrawn polyester filaments depending on maximal draw ratio (drawing temperature 20° and 70°C).

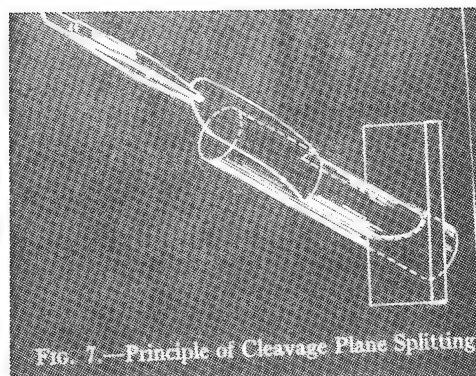
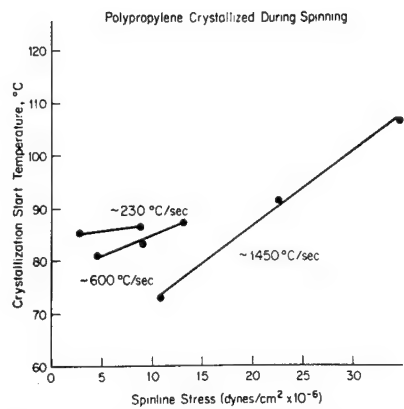
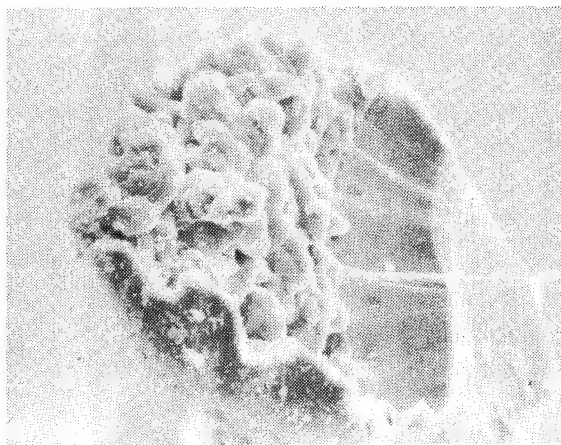
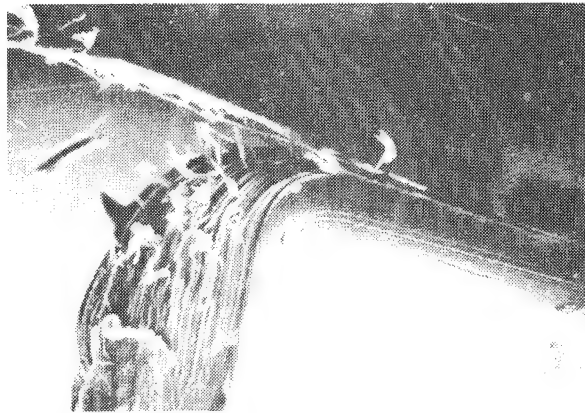
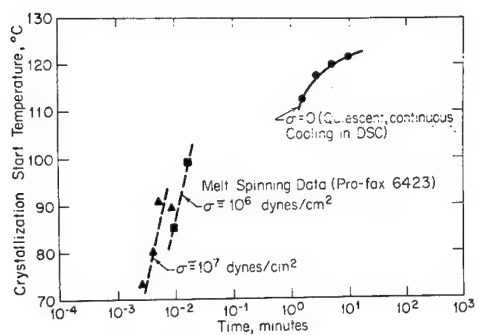
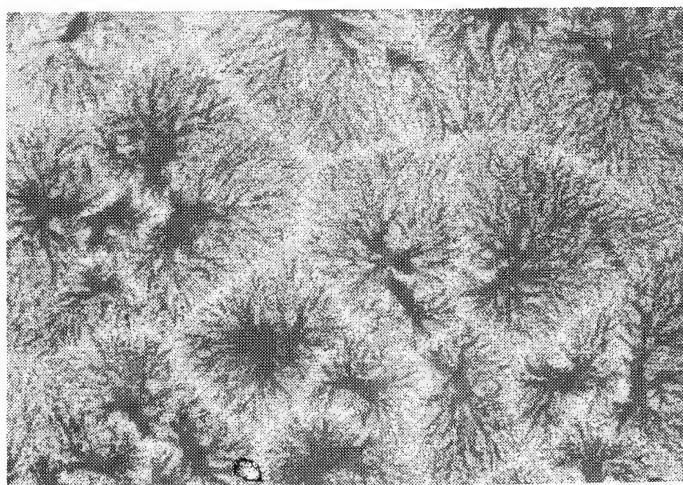
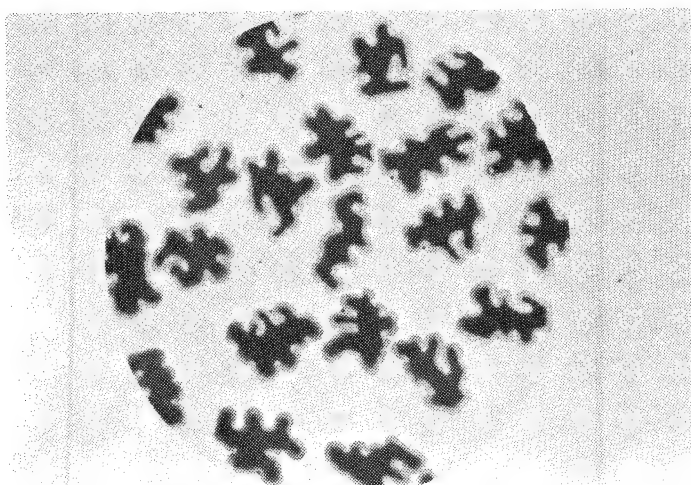


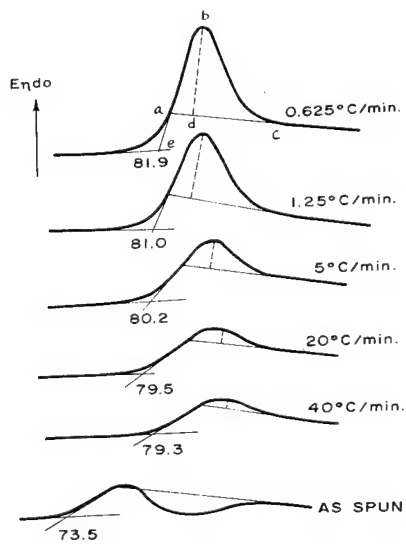
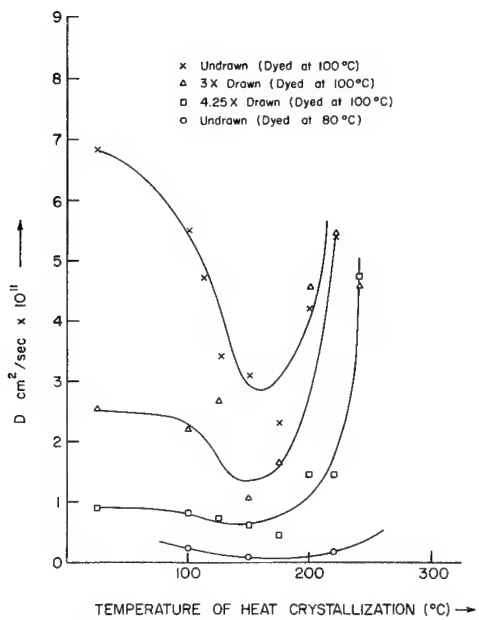
FIG. 7 — Principle of Cleavage Plane Splitting.



Crystallization start temperature versus spinline stress for polypropylene.



Continuous cooling transformation curves for isotactic polypropylene showing a comparison of melt spun and quiescent results.



Effect of cooling rate (thermal history).



EFFECT OF EXTRUSION PROCESSING PARAMETERS ON PROPERTIES  
AND STRUCTURE OF EXTRUDATE

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## Introduction

Plasticating extrusion is the process of applying heat and pressure to a resin feed stream to obtain a molten polymer. This melt is forced through a die to produce shapes continuously in various forms such as: 1) unsupported film, sheeting, tubes, and other profiles; 2) film for coating paper, metal foil, and other substrates and 3) coating electrical wire and cable.

The extrusion process is applied to thermoplastic polymers which can be melted and solidified repeatedly. The process is continuous in nature and the objective is to meter (deliver) a homogeneous melt at a constant controlled rate to the die. The die imparts the desired shape to the extrudate. When it emerges from the die the extrudate is cooled to set the imparted shape. The final properties of the extrudate depend on the processing conditions in the extruder and the post extrusion operations after the melt leaves the die. One can divide the properties of the product into those which are dependent on the quality of the melt and those which depend on the post extrusion operations after the melt leaves the die. The quality of the melt can be measured as follows(1):

1. uniform mass flow rate
2. uniform composition
3. uniform temperature

In turn the above criteria are dependent on both the extruder design and the operating conditions.

The finished product quality is a function of post extrusion finishing which is primarily concerned with heat transfer and extensional flows after the melt emerges from the die. The product quality may usually be categorized into two or three general classes. The first of these might be called aesthetic in nature, as evidenced by visual appearance factors such as roughness, gloss, haze, waviness and streaking. A second is classified as dimensional, meaning the ability to maintain size control to close dimensions. A third is functional in that the product must meet certain physical, chemical or performance test specifications.

#### Uniform Mass Flow Rates

Variations in output can be attributed to screw design, die design, resin (solid and melt) properties and processing conditions. For proper performance of the extruder the extruder must have a solids feeding rate which is greater than the melting rate and the metering rate. McKelvey (2) has stated this relationship mathematically as

$$r_1^* \geq r_2^* \geq r_3^* \quad (1)$$

where  $r_1^*$  = unrestricted solids transport rate

$r_2^*$  = unrestricted melting rate

$r_3^*$  = unrestricted pumping rate

When Equation (1) holds one obtains a stable operation. In order to insure the conditions of Equation (1) a number of specialty extruder designs have been developed. Kruder, Nichols, and Ridenour (3) provide an excellent review paper concerning the interrelationships of feeding, melting, mixing and pumping in the total

extrusion process. The discussion below is based on their paper.

### Extruder Feeding Alternatives

1. Extruder feed throat designs  
Round or rectangular of various dimensions, sometimes offset for tangential feed.
2. Gravity Feed Alternatives  
Conical or rectangular feed hoppers of various dimensions, sometimes equipped for vibration or agitation to facilitate deaeration or prevent bridging.
3. Solids Force Feed Stuffers  
Most commonly a vertical screw rotating in a short cylindrical exit at the bottom of a conical feed hopper inserted into the extruder feed throat. The vertical stuffer screw may be of variable dimensions and rotate at variable speed. Above the screw may be auxiliary compacting spirals and an anti-bridging agitator. Also, the cylindrical exit section may be either smooth or grooved, with or without cooling.
4. Plasticated or Melted Feed Applications  
Sometimes gravity feed where viscosity is not limiting. Otherwise use a stuffing pump or another tandem extruder to feed either under pressure or vacuum.
5. Internal Feeding Auxiliaries  
For materials with low coefficients of friction can use grooves or rifling in the feed throat and rear barrel to develop rapid pressure build up in the screw channel. Also, can use cuts or notches in screw to pull or masticate ribbon type feeds where appropriate.
6. Controlled Starve Feeding  
Either volumetric or gravimetric metering of a component or feed stream into feed throat. This feeding method is independent of screw speed (4).

### Melting, Mixing, Dispersion & Melt Pumping Alternatives (4,5,6,7)

1. Conventional Screws  
These screws are usually square-pitch single flighted with either a single or a two stage metering section. The design can be extended to more stages, either vented or non-vented, with other pitch angles, and/or multiple channels covering a wide range of section lengths, compression ratios, and meter depths. One can also use tandem extruders, either the "L" design or two separate extruders.
2. Localized Mixing Sections  
These sections are usually no more than 4 L/D in length. They are an integral part of the screw. The sections are incorporated at strategic locations for optimizing the screw's functions. The following types are available for localized enhancing of melting, mixing, and/or dispersion.

Union Carbide Mixing Section  
 Egan Mixing Section  
 Advanced Spiral Barrier  
 Reverse Flights and/or notched flights  
 Dulmage Interrupted flights  
 DIS mixing plug section  
 Rings or mixing pins  
 Slotted ring or undercut ring  
 Planetary intermeshing "Gears" or roller bars  
 Mixing teeth rings  
 Shear-cone or smear head  
 Intromix in-line static mixer

### 3. Longer Sections for Melting Promotion

In many cases more effective heat transfer is realized if one separates the melt from the solid bed or plug. This is also beneficial for heat sensitive materials where the overall processing temperature history is reduced. The following screw designs were listed by Kruder et al:

Maillifer Melt Separation Screw  
 "MC-3" (Barr) melt separation screw  
 Uniloy "Maxmelt" separation screw  
 Prodex Single and double flighted wave screws  
 Feedscrews, Inc., "Efficient" screws  
 Werner & Pfleiderer Twin Channel Compounding Screw

### 4. Motionless Mixers(8,9,10,11,12,13)

If after going through the extruder the melt is still non-uniform in either temperature or composition, the melt may be homogenized with motionless mixers. These units are in line devices which are placed between the tip of the screw and the die. The devices have no moving parts. It is the flow of fluid through the mixing devices which causes the mixing action. The concept of these mixers is to subject the material to a prescribed manner of flow. The purpose of the flow pattern is to divide, reorient, and recombine the stream so that components are randomly distributed throughout the fluid on some arbitrary scale of examination. The major commercially available units are:

#### 1. Kenics Thermogenizer

$$L = 2^E$$

#### 2. Ross ISG

$$L = 4^E$$

#### 3. Koch Mixer

$$L = \frac{m(2m)^{E-1}}{2}$$

where E = number of elements

L = number of layers

m = number of plates/element in Koch mixer

These mixers can reduce radial temperature variations to less than  $\pm 2^{\circ}\text{F}$  over a 2 inch diameter. The uniformity in temperature controls the melt viscosity and leads to a uniform product. Variations in composition are also blended out. Usually the amounts of color concentrate, fillers or other additives are reduced without loss in properties due to the uniformity of the melt composition.

#### Melt Flow in the Extruder Die

A second phase in the extrusion process concerns the flow of the melt in the die. For example a flat film sheet die has to perform a number of tasks which may be listed as (14):

1. Forming a molten web out of its total lip width combined with a complete material filling of the lip gap and locally exactly the same material velocities at the die exit.
2. Forming of a molten web of ultimate thermal and compositional homogeneity along the lip width, i.e. totally molten material of constant temperature and even additive distribution at the die exit.

In order to satisfy the above listed requirements the extruder and die must perform according to a prescribed set of operating conditions (14). The extruder has to show throughput characteristics which are free of surging, little affected by head pressure variations, and which are reproducible. Furthermore, the material delivered by the extruder must be homogeneous (both thermal and composition). Methods to accomplish this have been described previously.

The sheet die has to have a flow channel designed according to rheological considerations with regard to material processed, processing temperature, and throughput level. The preparation of extruded film is perhaps the most critical extrusion operation where the processing parameters can influence the final properties.

Incomplete melting will show up as an apple sauce effect in the melt extrudate. This in turn will cause optical distortion, local surface roughness and flow lines. Incomplete dispersion of color concentrates will show up as streaking. The incomplete melting and filtering of high molecular weight gels causes fish eye imperfections in the film. Flow in the die will lead to molecular orientation in the machine direction whereas the blow up ratio will cause orientation in the transverse direction. Orientation is a function of time and temperature. High shear rates and sudden chilling lead to maximum orientation. The molecules relax as a function of time. Temperature increases the molecular mobility and more relaxation can take place in a shorter period of time, i.e. the annealing time decreases with increasing temperature. Orientation can be physically correlated with birefringence studies for semicrystalline polymers. It can also be related to tear and impact strength. A study by Bakerdjian (15) indicates that film properties depend on the orientation balance subject to the above described time temperature relationship. He found that film properties such as dart impact strength, Elmendorf tear strength and puncture resistance are markedly affected by the degree of orientation balance imparted to HDPE film during fabrication. It was found that the die gap when varied within narrow ranges, does not affect film orientation balance to any significant extent. Blow ratio, however, has a very large influence on orientation balance and related film properties. High blow ratios consistently enhance film orientation balance and film toughness and tear characteristics. The results further indicate that film birefringence is a convenient quantitative measure of orientation balance and a

good indicator of film toughness and strength. A more theoretical analysis and measurement of the strain history in film blowing was carried out by Farber and Dealy (20). They also found that the balanced orientation as evidenced by shrinkage in the machine and transverse directions gave the best impact resistance.

The above described film blowing experiments indicate that the thermal history of the extrudate combined with extensional flow outside the die significantly influence the final properties of the extrudate, particularly profiles with thin cross sections. Thick cross sections are little affected because the polymers as a family have very low thermal conductivities ( 0.1 to 0.3 cal/g-cm<sup>2</sup>-°C-/cm) and the temperature profile changes very slowly with time. McKelvey has shown that density of a polymer is a function of both cooling (or heating) rate and temperature (17). The temperature determines the rate at which molecular orientation disappears which has been imposed during flow. As long as the temperature is uniform the time of relaxation will be constant provided the film thickness and degree of orientation are the same. Thin sections cool faster and more orientation is frozen in.

#### Developments in Blown Film Extrusion

In extrusion the length of the take off system is dictated by the time it takes to cool the extrudate. Semi-crystalline polymers pose a special problem in post extrusion forming since the latent heat of crystallization influences the temperature profile. Muzzy and Bright (16) have pointed out that one must consider the crystallization kinetics in order to predict the thermal history.



These authors have modeled the process by relating transport and thermodynamic properties to the growth of spherulites.

The properties of blown film can be markedly improved if the cooling rate can be increased. A number of attempts have been made to cool the bubble from both sides. Gregory(18) has reviewed the state of the art of internal bubble cooling and lists the following advantages:

1. Improved rate performance from blown film die.
2. Better film optical properties induced by the more rapid quench.
3. Improved surface optics such as haze by removing condensable volatiles from the bubble interiors of some systems.
4. Fewer film blocking problems inherent with high production rates.

Egan machinery Company (19) reported on a new system for internal bubble cooling of blown film. Egan claims an improved system consisting of an internal air ring, special die and rotator, and a control system for bubble diameter which regulates the flow of air in and out of the die.

#### Slit Film and Monofilament Extrusion

Scheiner (21) has given a recent review of the slit film and monofilament extrusion business. He found that the business had undergone a shakeout period over the last 10 years but that the annual growth rate should be about 10 percent/year. In both slit film and monofilament production major problems are caused by a lack of uniformity or consistency of product. Tape breakage usually is a major problem. The breakage occurs in the orientation

oven but is normally not the fault of the oven or orientation device. Residual water carryover on film is one cause; other causes are varying melt flow, unhomogenized melt, and high molecular weight resin leading to gels and weak spots in tape. Melt quality is most important. Usually screws with 24:1 L/D or greater are used. The compression ratio should be 3:1. The wave screw for polypropylene tapes gives good melt temperature uniformity and pressure uniformity. Die design is important since it is hard to make die heater adjustments or die lip adjustments for problems that occur in the die flow itself. Most polypropylene tape is made by the water quenched film process. PP blown film is a difficult and complex operation that requires special cooling rings. Chill roll cast film gives more uniform film thickness ( $\pm 1\%$  vs  $\pm 5\%$  on water quench). Since the chill roll cools from one side only the final product may be lower in tenacity. Monofilament extrusion the final properties are again imparted by orientation outside the die. Usually a two stage draw is used for better control and higher quality product. Scheiner believes that the toughest problem in monofilament work is the take-up of each individual strand after it is extruded and preventing these strands from contacting each other, at least until after quenching.

#### Orientation in Polystyrene Foam Extrusion(22)

Polystyrene extruded foamed film is used in many packaging applications. An important factor in successful utilization of the foam extrudate is the control of molecular orientation in the cell wall of the foam. A paper by Collins and Knaus reviews the process and the control of final foamed film properties as

a function of cell geometry. The orientation is essentially biaxial for film and sheeting with additional orientation in the final forming phase into a finished product. The film is produced in a tubular die and then passed <sup>over a</sup> sizing mandrel and cooled. Linear orientation is controlled by relating velocity of the polymer through the die lips to the take off velocity. The lateral orientation is controlled by the relation between die lip diameter and sizing mandrel diameter. A third orientation takes place during thermoforming. The cells undergo a post expansion during forming accompanied by a reduction in density and an elongation of the cells in the thickness dimension of the sheet.

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## BLOW MOLDING

### Rheological Analysis

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The purpose of this paper is to examine the effect of resin rheological properties on its processing behavior in blow molding equipment. Interactions between resin properties and processing parameters will be discussed, in particular: shear sensitivity, melt strength, die swell kinetics and melt flow instability. Special emphases will be placed on the high speed reciprocating blow molding of polyethylene where the material is often subjected to shear rates which are well above the onset point of melt instability and on the blow molding of high molecular weight resins.

THERMOFORMING PROCESSING MATERIALS PROPERTIES AND THEIR INTERRELATION

PRESENTED AT THE

SOCIETY OF PLASTIC ENGINEERS

ENGINEERING PROPERTIES DIVISION

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MOORE BUSINESS FORMS, INC.

Any successful product is a combination of material choice, product design and processing. It is not the intention of this paper to select materials for an application. I assume you have already made this selection. This paper is meant to indicate those thermoforming variables you should be aware of to allow the successful processing of this material to produce a quality part.

#### REVIEW OF THE THERMOFORMING PROCESS:

The thermoforming process takes a sheet or film of thermoplastic material, heats it to its softening temperature then by means of mechanical or pneumatic pressure forces the sheet onto or into a mold. After reproducing the detail of the shape and texture of the mold, the sheet is cooled to a temperature where the part will not distort upon release from the mold. The sheet from which the part is made is always larger than the part produced. Trimming of the part from the sheet has been considered a post operative function of thermoforming, but I consider it a necessary step to produce a part. Production rates on thermoforming are directly associated with the starting and finished plastic material thickness and the size of the part produced. Commercial production rates vary from 1500 parts per minute to 1 part every 15 minutes.

In the thermoforming process there are four major areas. First area is the plastic raw material itself. Second will be the heating of the plastic. Third will be the forming into a part and the fourth will be the removal of the part from the sheet of plastic material. All four together will yield a finished part.

#### PLASTIC RAW MATERIAL:

In the thermoforming process, extruded plastic sheet is the raw material. This differs from injection molding, rotational casting and blow molding which utilizes a 1/8" cube of plastic material, as supplied from the chemical companies. Extruded sheet as the raw material offers an advantage to the process. Definition of texture, color and physical properties are known prior to converting them to a finished part. This is a quality control step not available in injection, blow and rotational molding. Because the plastic material has been processed into a sheet, it will also have the influences of the extrusion process, i.e., orientation of the sheet, variation in material thicknesses. Currently being processed in the United States are the polystyrenes, ABS materials, rigid and flexible vinyls, polyethylene, polypropylene, acitates, polycarbonates, nylons, urethanes and the polyesters. It is safe to say that any thermoplastic material that can be formed into a sheet by extrusion or calendering can be thermoformed.

From the physical properties list of a particular raw material, we find tensile strength at break, elongation at break, Izod notch impact, thermoconductivity, thermoexpansion and continuous service temperature. Each one gives a control definition to the variables of the thermoforming process. Information not commercially available to us, but very important in setting our process variables are molecular weight, the molecule itself, the molecular weight distribution, hot strength and melt zone.

Commercial thermoforming equipment provides for heating the thermoplastic material, forming it into a part, and trimming the part from the plastic sheet.

### HEATING

Energy for heating the plastic sheet is normally supplied by electrical resistance heaters or by the combustion and natural gas. With the current shortage of natural gas, this is virtually an obsolete heating approach. Using electrical resistance heaters in the equipment, the heat is supplied to the plastic sheet by conduction, convection and/or radiation. Conduction heating of plastics is not a widely used method due to the problems of the adhesion of the plastic to the heating surface as the sheet reaches its forming temperature. Convection is the most widely used method.

From appendix #1, Physical Properties of Plastic, we can now see the thermoconductivity values of one plastic verses another, and we can see that this compared to the good conductors of heat, like aluminum, are relatively low. Therefore the transfer of heat throughout the sheet is a relatively slow process. Convection heat input into the plastic sheet is dependent on the rate of acceptance of the heat by the plastic. If the input rate of heat is greater than the acceptance rate, surface temperature builds up on the plastic resulting in scorching or degrading of the material surface.

Electric resistance heating elements admit radiation in the 900° to 1200° F surface temperature. Chart #1 is a plot of wavelength emission verses the surface temperature of electrical resistance heaters. Efficiency of the radiant heat is dependent on the distance between the heating elements and the plastic sheet itself. It is necessary to generate a uniform radiant energy since a uniform rate of heat input to the plastic is essential.

Most commercial machinery heats plastic sheets by both convection and radiation. The best processing control is with the plastic sheet at the same temperature overall. This does not allow hot or cold spots in the plastic material. Although the basic polymer sets the thermal conductivity and the infrared absorption rates, the color and filler in the plastic and the surface sheen or texture can radically change the total heating rate.

### FORM

Next physical section of the thermoformer is the Form Stations. The form station of any thermoformer has two platens; one mounted over the other and mechanically actuated together in a part. Pneumatic, hydraulic and/or mechanical actuators create the required forces on the platens. Onto the platens to the form stations are bolted lightweight aluminum water cooled tools that can either be male or female molds. Part design and desired material distribution of the plastic will predetermine whether a male or female tool is used.



Mechanical devices called plug assists are usually employed with male and female tools and help distribute the plastic material into mold areas during the forming process. The ability to control material thickness on an area of the part, allows for control of part weight while maintaining functional requirements.

There are 9 different processes for thermoforming. Reference Appendix #2. Figure 5 of that table is plug assist vacuum forming. By comparing one method against the plug assist vacuum the variation in process is obvious. In the plug assist vacuum forming, once the sheet has been heated and brought across the mold cavity, a plug assist shaped as to the mold cavity, but slightly smaller, is plunged into the plastic sheet and prestretches the material. When the plug platen has reached a desired closing point and delivered the material to a particular position relative in the cavity, vacuum is applied on the mold cavity. An optional process is to apply pressure on the plug assist side of the sheet. The areas that the plug touches on the form part, usually create thicker areas due to the chilling effect of this contact. Consequently, plug design is most significant as to its shape configuration as well as its own temperature control.

#### TRIMMING:

The last physical step in thermoforming process is the removal of the part from the sheet. There are two basic approaches to removing the part from the sheet under production conditions: First is steel roll die trim and Second is match metal die trim. Usually trimming the part is done as a separate operation to the forming and is done in separate sections or pieces of equipment. Steel rule die trimming is employed on lower volume production runs and where dimensional tolerance demands are relatively low. Steel rule die consists of a steel band approximately 1/16" thick and 1" high with 1" ground to a knife's edge. The blade is mounted in a plywood frame allowing the 1" to extend at a 90° angle to the plywood. The forces in trimming are applied to the steel blade base in the plywood and transmits the forces to the plastic at the sharpened edge. The plastic is trapped between the blade and a reinforced stainless steel facing. Force causes the blade's sharpened edge to enter into the plastic.

In steel rule die trimming, 100% separation of the part from the web will leave the part in the trim die. This is undesirable since collection of the part then is difficult. A 99% separate is desired which allows part collection of the parts. The life of a steel rule die is dependent on the application, the polymer and trim complexity. Normally one hundred thousand to a million parts representative of the life of a steel rule die. For parts in excess of one million, match metal dies are usually employed. Match metal trimming also allows higher trim accuracy. Match metal trim dies, hardened steel punches and dies are mounted in a die set with four post guiding. The die post guarantee alignment of the punch and dies over a prolonged production run. The tolerance between the punch and the dies is dependent on the material being trimmed.

In the match metal die trimming, the part is pushed through the opening of the die, collection of the parts is then possible by a variety of mechanical devices, conveyors, counting, bagging systems are employed to eliminate manual handling. One operator usually runs more than one forming line.

#### VARIABLES FOR HEATING:

Now that you have a general idea of the thermoforming process, let's talk about the various process stages, the heating, forming and trimming and how they relate to the physical properties of the raw materials. Physical properties of interest to us will be tensile strength at break, percentage elongation at break, izod notch impact, thermoconductivity, specific vicat softening point, continuous service temperature, linear thermal expansion. These values have been collected from the International Plastics Selector of 1977 and are found in our appendix #1. To allow the form station to mechanically and/or pneumatically reshape the plastic raw material, it is necessary to raise the temperature of the plastic so that the physical forces available in the form station will exceed the tensile strength of the material. The question is, how much energy will be required and how much time will be necessary to heat the plastic mass to a specific temperature where we can process material. From the appendix and physical properties an item called vicat softening point is a key. ASMD 15 - 25 test methods, the vicat softening point is the temperature in which moderate pressure will allow an indentation foot to penetrate the plastic by 1MM. This is the start of the physical working zone of the plastic. Tensile strength verses temperature virtually does not exist for the thermoplastic raw materials. Using the tensile strength at room temperature, we can project the tensile strength verses temperature curve. See Graph #2. Tensile strength, at room temperature, will virtually decrease to 0 as the melt point of the plastic is reached. Prior to the melt point though, will be a softening point that the plastic has indicated by the vicat softening test. Somewhere between the melt point and the softening point will exist a working zone which will allow us to form the plastic. This we'll refer to as it's working zone and the tensile values in the work zone will be referred to as the hot strength of the plastic. The rate of decrease of tensile properties in this work zone can be approximated by the melting curve of the polymer verses the caloric input. We refer you now to graph #3. In the melt curve, polymer verses caloric input, we can make a number of observations. One is that having a plastic material of single molecule at a particular molecular weight, we would expect a very sharp change in the melting curve as it reaches the melting point. This is probably best reviewed from experience of nylons injection molding grade of materials, where at 490°, the material is virtually impossible to inject into a mold, but at 500°, it runs like water. Today, the thermoforming plastic polymers virtually are composite materials of molecules as well as a variety of molecular weights, so that the melting curve is not as we've shown. It would more look like the next graph which is #4.

The end result of a combination of different molecules and different molecular weights is a considerably broader melting zone as well as a wider work zone for our forming process. The ABS materials are probably the best example of a combination of molecules and a variety of molecular weights and is represented in the thermoforming process as one of the easier materials to work. This is due to wider work zone and the good hot strength for forming. An example of a very sharp melting point material, going back to graph #4 is the homopolymer of polyethylene at a particular molecular weight. This is specifically one molecule, one molecular weight that gives you a very narrow range of molecular weights. The result is that for thermoforming, you will have a very narrow workzone for processing.

Everything that we have said so far has had to do with virgin raw materials. We can change the molecular weight distribution by the addition of reground material with virgin material. In the thermoforming process, 20% to 40% of the plastic sheet is reground for reprocessing. In the regrinding and reprocessing, the plastic material experiences a second heat influence and the possibility of polymer degradation exists. Polymer degradation actually changes the molecular weight of the polymer by decreasing its molecular weight or cross-linking and increasing its molecular weight. Different polymers react differently into this heat history, and because of it, some materials can be readily reground and in high concentrations be put back into the virgin material for re-extrusion while others can only be processed one or two times and be put back into the virgin material for re-extrusion in very limited quantities. An example of that would be polyurethane materials.

Another way to change the working zone and hot strength of the plastic is by orienting the sheet on extrusion. Orientation is the stretching of the plastic in either direction of extrusion or at 90° to the extrusion direction as it is being processed through the pull/cooling rolls. This gives the strain in the plastic, put in during extrusion, which is not relieved until the temperature at which it's been put in has again been reached. In our thermoforming process, we do not normally reach these temperatures again and 100% relief is orientation. The result is that the material will have a latent rubber band tensile strength effect within the plastic sheet. The result is a higher tensile strength in the working zone for our processing. This also gives the physical part that we're producing higher strengths. In a thermoforming process, it means that our ability to produce the part requires greater forces.

Orientated sheet virtually eliminates straight vacuum forming as the approach to produce a part. Straight vacuum forming has only 14.0 pounds per square foot pressure available to shape the plastic during forming and the orientated sheet will have hot strength above this. Other items that should change the tensile property versus temperature curve will be color concentrates and/or fillers that have been added to the plastic material.

From the previous discussion on heating we know that heat is put into the plastic sheet by convection, conduction and radiation. From our chart of physical properties, we find the thermoconductivity of the material and the specific heat, this along with softening point will allow us to approximate the time required to heat the plastic. Unfortunately, we can't calculate this time to heat the sheet due to the fact that we do not have the radiant energy absorption capability of the plastic. Fortunately from experience, we know that high impact styrene, ABS and PVC have overall heat input rates of approximately 2 mil. per second. For 30 mil. ABS stock, 15 seconds of heating is required. For polyethylene and polypropylene, the rate of heat acceptance is approximately 3-3½ mils per second. These rates change with the color, fillers and textures of the materials, and our quoted values are approximate.

Referring back to the physical property charts, the thermoconductivity of ABS versus polyethylene, we can see that it's almost at 1:2 ratio allowing the polyethylene to transfer heat internally twice as fast. Because of this, the heat rate of the polyethylene material is expected to be considerably higher and it is an experience. Also the thermoconductivity will be important to us after we have formed the part, since it must cool the part on the mold for releasing it. In this cooling process, also specific heat will be significant.

You will find that in comparing specific heats from the materials that the polyethylene have almost twice the amount of heat to remove as with the ABS materials, that will be a processing disadvantage.

In the forming process we will have to make a choice of the various thermoforming processes available to us as well as mold temperatures, plug assist temperatures and the sheet temperature work zone that we prefer to be in for processing. When we talked about the tensile strength versus temperature curve and talked about the work zone of the plastic and the hot strength of the plastic, we did not mention that the work zone was dependent on the pressures used for forming the plastic in the thermoforming process. Utilizing the straight vacuum force to pull the plastic sheet onto the mold, fourteen pounds of pressure is available to form the part. By utilizing the pressure push on the plug side of the sheet then the forming we can multiply the pressures available to 80 or 90 PSI, depending on the tool capacity as well as the form station capacity. By the addition of pressure we can lengthen the work zone in which the plastic can be formed. Other techniques can also utilize match metal tooling which would exert even greater forces than the 80 or 90 PSI available. This approach for forming is desirable for polymers which have an extremely narrow molecular weight of a particular molecule. So from the work zone and hot strength of the material, the type of process used to manufacture the part can be tentatively selected. Material distribution has to be considered before the final process selection is made. With the process defined, we can utilize the physical property charts to lock in on mold temperature, as well as plug assist temperature. Our continuous service temperature, the plastic listed physical properties, is the temperature at which the plastic material will not distort

on use. This is the maximum temperature that the mold is to be controlled during the cooling of the plastic part and prior to its removal from the mold. Plug temperatures then will be very close to the vicat softening temperature. It is desirable that the plug assist does not significantly chill the plastic material when it touches it. This means that we must maintain the plug assist temperature in work zone. This is approximately the area of vicat softening point. The only other thing to consider in the forming process is the timing to cool the plastic to the point where it can be removed from the mold. This is dependent on the temperature which the sheet is supplied to the mold, the material thickness. The thermoconductivity of the plastic material and the specific heat of the material. We know that the continuous service temperature point is the upper temperature that the part should be at prior to removal from the tool. It is to be remembered that the plastic material on the mold is always thinner than the starting material of the sheet.

In the thermoforming, the plastic sheet is always larger than the part or parts that we've generated. It is therefore necessary to remove the part from the plastic sheet as the final step in thermoforming. In this removal process called trimming, it is therefore desirable to anticipate the forces required to remove the plastic material from the web and will be dependent on the material thickness, the tensile strength of the material and the izod notch impact effect. These will then be able to allow us to predict the number of lineal inches that we can trim at any one time. From the physical properties chart, we can then obtain the tensile properties of the plastic that we have selected and the izod notch impact.

Extending the work zone is probably one of the best approaches for production. If the zone of processing is too narrow, then scrap rates will run exceptionally high. If we can extend the working zone, we can then offer a production process with more latitude and better production rates with lower scrap.

In the trim station, we are going to remove the form part from the web or the plastic sheet. The forces required to trim that part is dependent on tensile strength of the material, the izod notch impact and the lineal inches of material to be trimmed and it's thickness. Tensile strength gives us the absolute values necessary to trim the plastic at room temperature. The izod notch impact gives us an indication whether the trimming force is required through the entire thickness of the plastic or whether a material fracture will occur as the force is applied. A fracture break reduces the tensile strength to trim the part. It results in a reduced force required from the theoretical calculatable values.

The percentage elongation at break gives a clue as to the manner in which the plastic breaks on it's separation from the sheet. In general terms, the higher the percentage of elongation, usually the higher the izod notch impact.

With low values for percentage of elongation and izod notch impact, a fracture break in trimming will result in lower than the theoretical forces. Therefore, to calculate the forces only from tensile strength and thickness and linear inches is not sufficient.

Using a match metal trim die, die clearance between the punch and the die is essential to be established prior to its manufacturing. Our percentage elongation and our izod notch numbers will indicate the tolerance between the punch and die. From experience, it has been found that styrene and ABS parts (with a relatively low percentage of elongation had a relatively low izod notch impact), require a die clearance of between 3 and 6 thousandths when the percentage elongation at break is high and the izod notch impact is high. The clearance between the die to accomplish the separation of the part from the plastic is considerably tighter. Half a thousandth to one thousandth is required for the polyethylene materials.

The edge finish required at the separation of the part from the plastic is dependent on the izod notch or the fracturability of the material. The lower the izod notch impact, the more fracture and irregularity of the fracture lines in the existing thickness, creating possibly an undesirable part at the trim line. This can be overcome by mechanically squeezing the trim line area prior to the trimming and reducing the thickness at the cut line.

One of the most important items to be calculated in the trimming is the size of the punch and die versus the tool size in the form station. The linear thermal expansion of the material and the temperature range in which the plastic travels will allow you to anticipate the change in size of the hot part versus the cool part for trimming.

With a trim press at a stated capacity and the forces required to trim one part from the plastic sheet, the number of parts to be separated per stroke can be resolved.

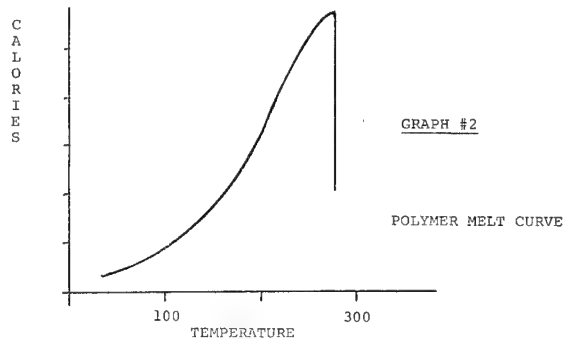
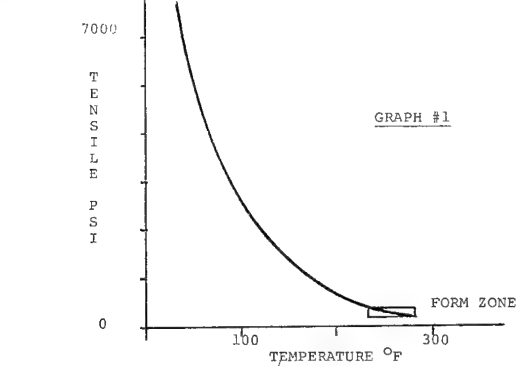
For plastic with low izod notch impact values, there must be a difference between the calculated forces per number of parts versus the capability of the press. Should the calculation show 10 tons of force is required per stroke, a 15 or 20 ton press should be utilized. If not, the impact of the punch into the plastic will send a significant shock wave through the press. This will shorten the life of the trim press.

Tonage can be reduced by stopping the punches and dies. In this way, not all the punch impact will be felt at the same time. The result is a distributed force over a longer time period. Putting shear on the punch will also accomplish the same result.

At this point we have covered the Heating, Forming and Trimming functions of thermoforming and natural properties.

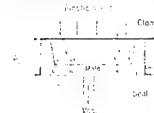
The facts and ideas presented in this paper are meant to outline and lightly cover some of the variables in the thermoforming process.

I've tries to avoid specific applications and I invite you to apply some of this information to your own applications and problems.



## Thermoforming techniques

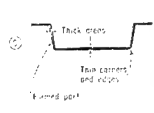
**Fig. 1: Straight vacuum forming.** The plastic sheet is clamped and heated. A vacuum beneath the sheet (A) then causes atmospheric pressure to push the sheet down into the mold. As the plastic contacts the mold (B), it cools. Areas of the sheet touching the mold last are thinnest (C).



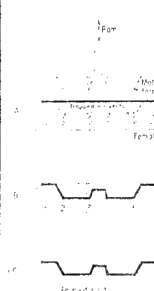
**Fig. 2: Drape forming.** The plastic sheet is clamped and heated (A), then draped over the mold either by pulling it over the mold or by forcing the mold into the sheet. When the mold has been forced into the sheet and a seal created (B), vacuum applied beneath the mold forces the sheet over the male mold. By draping the sheet over the mold, that part of the sheet touching the mold remains close to the original thickness of the sheet. Side walls are formed from the material trapped between the top edges of the mold and the custom seal area at the base. Final wall thickness variations are shown in the drawings (C).



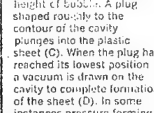
**Fig. 3: Matched mold forming.** Matched molds of wood, metal, plaster, epoxy, etc., can be used to press the sheet to shape. The heated sheet may be clamped over the female die (A) or can be draped over the mold force. As the mold closes it forms the sheet (B). Mold walls allow trapped air to escape. Clearance between the mold force and cavity of the mold depends upon tolerances required in the part. Excellent reproduction of mold detail and dimensional accuracy can be obtained from matched mold forming, including lettering and grained surfaces. Material distribution of the formed part (C) will depend upon the shapes of the two forms.



**Fig. 4: Pressure bubble-plug assist vacuum forming.** After the plastic sheet is heated and sealed across the female cavity (A), air is introduced from the mold cavity and blows the sheet upward into a bubble (stretching it evenly) (B). Normally an electric photocell is used to control height of bubble. A plug shaped roughly to the contour of the cavity plunges into the plastic sheet (C). When the plug has reached its lowest position a vacuum is drawn on the cavity to complete formation of the sheet (D). In some instances pressure forming air is also used in this process.



**Fig. 5: Plug assist vacuum forming.** After the plastic sheet is heated and sealed across the mold cavity (A), a plug, shaped like the mold cavity but smaller, is plunged into the plastic sheet and pushed stretching the material (B). When the plug reaches its lowest position (C) a vacuum is drawn on the mold cavity to complete formation of the sheet. Wall thickness can be varied by changing shape of the plug (D). Areas of the plug pushing the sheet first create thicker areas due to the chilling effect. Consequently, plug design is most important factor in determining just what the geometry of the formation being produced by this technique will look like.

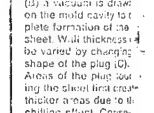


**Fig. 6: Matched mold forming.** Matched molds of wood, metal, plaster, epoxy, etc., can be used to press the sheet to shape. The heated sheet may be clamped over the female die (A) or can be draped over the mold force. As the mold closes it forms the sheet (B). Mold walls allow trapped air to escape. Clearance between the mold force and cavity of the mold depends upon tolerances required in the part. Excellent reproduction of mold detail and dimensional accuracy can be obtained from matched mold forming, including lettering and grained surfaces. Material distribution of the formed part (C) will depend upon the shapes of the two forms.

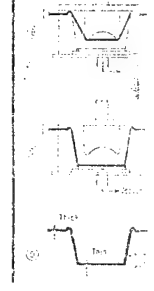


Fig. 6: Matched mold forming.

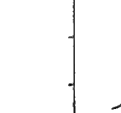
**Fig. 7: Plug assist vacuum forming.** After the plastic sheet is heated and sealed across the mold cavity (A), a plug, shaped like the mold cavity but smaller, is plunged into the plastic sheet and pushed stretching the material (B). When the plug reaches its lowest position (C) a vacuum is drawn on the mold cavity to complete formation of the sheet. Wall thickness can be varied by changing shape of the plug (D). Areas of the plug pushing the sheet first create thicker areas due to the chilling effect. Consequently, plug design is most important factor in determining just what the geometry of the formation being produced by this technique will look like.



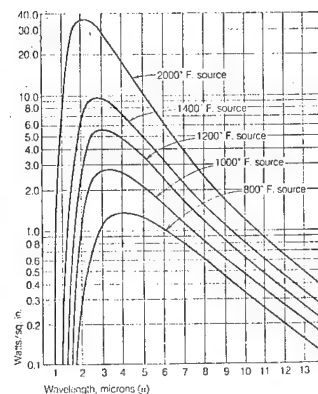
**Fig. 8: Matched mold forming.** Matched molds of wood, metal, plaster, epoxy, etc., can be used to press the sheet to shape. The heated sheet may be clamped over the female die (A) or can be draped over the mold force. As the mold closes it forms the sheet (B). Mold walls allow trapped air to escape. Clearance between the mold force and cavity of the mold depends upon tolerances required in the part. Excellent reproduction of mold detail and dimensional accuracy can be obtained from matched mold forming, including lettering and grained surfaces. Material distribution of the formed part (C) will depend upon the shapes of the two forms.



**GRAPH #3**



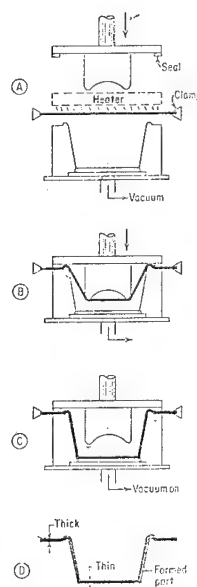
**IDEAL POLYMER MELT CURVE**



Distribution of radiant energy vs. wavelength. Most thermoplastics absorb heat in 2 to 6 micron range more efficiently than shorter wavelengths.

## TYPICAL PHYSICAL EXTRUSION PROPERTIES

	TENSILE STRENGTH (YIELD)	ELONGATION (BREAK)	IZOD NOTCH	THERMOCONDUCTIVITY	SPECIFIC HEAT	VICAT SOFT	CONT. SERV.	LINEAL THERMAL EXPANSION
	ASTM D-638	D-638	D-256	C-177	D-696	D1525	C-351	D-696
	Lbs./sq.in.	%	Ft./lbs/in	BTU.in./Hr.Ft. <sup>2</sup> °F	BTU/Lbs °F	°F	°F	10 <sup>-5</sup> in/in. °F
POLYSTYRENE G.P.	6400 PSI	1.5%	0.25					4-5
	4200	30	1.25	1.1 - 1.4	.30	193	145	4-5
	3300	43	2.2			210	150	4-5
ABS	6600	25	1.0			217	122	
	5100	40	4.0	1.3 - 1.7	.35			5-6
	4400	50	6.0					
POLYCARBONATE	9500	110		1.3 - 1.5	.27	315	250	3
POLYESTER	8000			1.1	.4			6-7
POLYETHYLENE	2600	700	6.0					
	4100	600	6.9	2.3 - 3.4	.5	250	180	7
	2000	500						
POLYPROPYLENE	1800	550			.55	210	140	11
	4800	5.50	2.1	1.4 - 1.5	.5	200	220	4.4
	4300	700	5.4					
POLYVINYLCHLORIDE	8500	50	.54			160-180		
	6400	30	2.0	1.16	.25			3.8
	3000	300						



MACHINERY AND METHODS



# REVIEW OF FORGING, STAMPING, AND OTHER SOLID-PHASE FORMING PROCESSES

by

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## ABSTRACT

In the last ten years, a considerable amount of work has been conducted on many different processes distinct from common plastics processing methods such as injection molding, extrusion, and compression molding. Perhaps the most common feature of these new techniques is that they resemble more the methods used for fabricating metallic materials in solid phase. However, there is a lack of uniform terminology. More importantly, the commercial acceptance and applicability of these processes have been slow.

This review attempts to summarize the important findings of the various publications and, whenever possible, to give information about potential or actual industrial applications. The purpose is to provide a brief overview of the state-of-the-art of the whole technology. Advantages, disadvantages, and the need for more work are pointed out as they relate to different processes and to crystalline and amorphous materials. A few instances where material was heated beyond the melting temperature by a small amount are included in the review when the processing method was similar to solid-phase forming.

## INTRODUCTION

In the last ten years, much work has been carried out on fabrication techniques for plastics which resemble more methods used commonly for metals. This paper will review publications that have appeared primarily in the last five years or so and also present some unpublished results. The two main categories

of processes considered are stamping and forging. 'Stamping' covers all of the techniques employed for sheet-type workpieces, whereas 'forging' refers more to bulk deformation. In each case, the principle and main advantages of the process are described, new information about the basic understanding of the process is reviewed, and new techniques or commercial applications of the process are pointed out. Finally, some of the other techniques are referred to briefly. The main thrust in this paper is to summarize key new information about processing and applications and to give an overall picture of the state of the technology.

'Solid-phase forming' is a general term applied to this whole category of processes similar to metalworking methods in which the main characteristic is that the work material does not have the fluidity to give rise to the type of fluid flow observed in processes such as extrusion or injection molding. In general, for amorphous materials, this means that the processing temperature is below the glass transition temperature and for predominantly crystalline materials, this means that it is below the crystalline melting temperature. In metalworking the term 'hot working' is applied when the processing temperature is above the recrystallization temperature of the material. In polymers, crystallization takes place in a range of temperature starting from the melting point to a temperature somewhat below that. Hence, the techniques discussed in this paper would generally fall in the category of 'warm-forming' operations. When the material is not heated--that is, it is processed at the ambient or room temperature--it is referred to as 'cold forming.' Additional definitions will be provided in the subsequent sections as they become necessary.

A comprehensive review of much of the published literature on solid phase forming up to early 1971 is given in Ref. 1. A brief overview of forging is presented in Ref. 2. These references, along with the additional bibliography in each, cover

well much of the early literature on the subject which, for the sake of brevity, is not covered in this paper. Table 1 gives glass transition temperature for some common amorphous materials and crystalline melting points for some common crystalline materials. These data derived from Ref. 3 will be useful in subsequent discussion of the processing conditions for various materials.

## STAMPING

### Principle and General Comments

Stamping is a broad term applied to all press working operations.<sup>(4)</sup> For stamping, the workpiece is generally in the form of a thin sheet of uniform thickness. Any stamping operation can be considered a combination of three basic types of deformation, Fig. 1. The simplest is the 'bending' in which the permanent deformation is restricted to a narrow portion around the bend and all the other parts of the workpiece are subjected to simple movement (not deformation). The second type of deformation is 'stretching' in which the biaxial strains on the surface are everywhere positive. Pure stretching occurs, for example, when the blank holder pressure in forming is high enough to prohibit any movement of the sheet past the blank holder. Since the surface area increases during stretching, it is accompanied by thinning of the workpiece. 'Drawing' is a deformation process in which the permanent deformation results in a positive strain in one direction in the plane of the sheet surface and a negative strain at 90° to the previous strain in the sheet surface. Such deformation can occur only when the blank holder pressure is low enough to permit sheet movement underneath it. In a pure drawing operation, the original surface area and the thickness of the workpiece are unchanged. When the depth of draw is higher than the minimum width of the formed shape, it is termed 'deep drawing.'

Important processing conditions in a stamping type of operation include design of the blank geometry in relation to the

geometry of the component to be made, blank holder design and pressure, lubrication, tooling geometry, and deformation speed. In warm forming operation the temperature of the preform is another important variable. Additionally, the ease or difficulty of forming (formability) is greatly influenced by the condition of the original workpiece (e.g., as-extruded vs. biaxially rolled) and its thickness. Some of the common defects in the stamping operation are wrinkling of the side walls or the blank holding area when the blank holding pressure is low, or splitting or unacceptable necking of the bottom of the formed shape when the blank holder pressure is high. Under any given conditions, the formability is generally expressed in terms of the limiting draw ratio (LDR) which is the ratio of the blank diameter to the diameter of a formed cup corresponding to the largest diameter of the blank which can be successfully formed into a cup (without any lip). It is generally accepted that a higher LDR is possible when the sheet is subjected to biaxial rolling prior to stamping. However, the reasons for this are not fully understood and this is a subject of continued study. Additionally, investigations have concentrated on the springback of the formed shape and the load required for stamping. Springback refers to the tendency of the formed shape to revert back partially or wholly to the original sheet configuration after releasing the load or when subjected to higher temperatures. Excessive springback causes poor fidelity of the formed cup to the desired configuration and is a major concern in a forming operation.

#### Basic Investigations

Many of the investigations in stamping have been for cold forming operations whereas the commercial applications have been more for warm forming.

Cold forming of plastics in drawing type of operation was investigated by Li et al.<sup>(5)</sup> for a variety of materials. They indicate that for a thermoplastic material to be cold formable,

the glass transition temperature of the polymer should be above the ambient temperature, the tensile elongation should be at least 30%, and the ratio of tensile to compressive yield strength should be at least 1.6%. Further, the sheet should not yield locally (neck) when strained in tension. Figure 2 from their work shows how the LDR can be estimated from a series of experiments with different blank diameters and blank holder pressures. The LDR corresponds to the ratio of the blank diameter at the point of intersection of the two curves in Fig. 2 to the punch diameter. The authors found that some of the polymers such as ABS and PVC may show good or bad response to forming depending on the exact formulation whereas material such as Nylon 6 and Nylon 6,6 show varied response depending on moisture absorption. The dry nylons show good formability and good shape retention whereas the wet nylons, although formable, show poor retention. The relaxation in shape (springback) of the cold formed cup when the cups are subjected to different test temperatures is shown in Fig. 3 and gives some indication of the amount of springback that can be expected from these materials.

On the whole, the above authors felt that the high springback and low heat distortion temperature of the cold formed items pose severe limitations to their practical usage. As for the local yielding during forming, it can be avoided by biaxial rolling of the sheet prior to forming. As a continuation of this work Prevorsek et al.<sup>(6)</sup> studied cold forming of laminates containing cross-linkable cores. A satisfactory construction was found in a composite consisting of a filled, glass-reinforced epoxy core sandwiched between two PVC/aluminum face sheets. The aluminum layer provides a barrier between the core and PVC and increases the resistance to heat distortion of PVC. The advantages of such composite forming were suggested to be the capability of fast forming of thermosets with functional and/or decorative surfaces, high modulus, and high heat distortion temperature as compared with all thermoplastic sheets.

Deep drawability of biaxially rolled thermoplastic sheets was the subject of an investigation by Broutman et al.<sup>(7)</sup> The authors attempted to apply the theory employed in sheet metal forming to forming of polymeric materials. In sheet metal working the anisotropy of the sheet is expressed in terms of normal and planar anisotropy. The normal anisotropy  $\bar{R}$  is defined as follows:

$$\bar{R} = (R_0 + 2R_{45} + R_{90})/4 \quad (1)$$

In the above formula,  $R$  represents the strain ratio obtained from a uniaxial tensile specimen with the subscript denoting the direction of testing in relation to a reference direction (normally the direction of rolling for sheets metals). The strain ratio is the ratio of the true strain in width direction to true strain in thickness. In sheet metal stamping, it is known that the higher the normal anisotropy ratio, the higher the resistance to thinning and hence the higher LDR of the material. The authors found that unlike metals, the normal anisotropy in polymeric materials is a function of the amount of elongation used in uniaxial tensile tests carried out to calculate the strain ratio, Fig. 4. Interestingly, the LDR for the polymeric materials was found to be related to normal anisotropy (Fig. 5) as in the case of the metals. The authors again observed that biaxial rolling of the polymers improves the LDR, especially for polycarbonate, and that this increase is related to normal anisotropy increase. The authors confirm that biaxial rolling can eliminate necking and stress whitening. Also, lubrication of the sheets reduces the forming load and thus a larger blank diameter could be drawn before fracture occurred. Thus, the LDR increased with lubrication. This differs somewhat from sheet metal working where unlubricated punch and lubrication on the lower side of the sheet are the conditions associated with the best drawability. The authors also found that biaxial rolling of the material changes the true stress-true strain curve for the polymer to that similar to a metal and it can be expressed

in the form of  $\sigma = k\epsilon^n$ . However, the strain-hardening exponent  $n$  did not appear to have any relationship with drawability.

Evans<sup>(8)</sup> also investigated the deep drawing process and developed analytical expression which enables calculating the LDR on the basis of only six experiments carried out using standard diameter blanks. His analysis allows for the elastic deformation of the blank during the forming process. The extrapolated values of LDR given by Evans seem comparable to those determined experimentally by Broutman et al.<sup>(7)</sup> His experiments suggest that von Mises' yield criterion gives reasonable accuracy in evaluating the stresses in the annulus of a cup during drawing. He suggests that any mechanical or chemical treatment which results in an increase in the ratio of the tensile stress at failure to compressive yield stress should increase the LDR of the material.

Miles and Mills<sup>(9)</sup> have critically investigated the application of the theory of deep drawing of metals to thermoplastics. They contend that the strong orientation hardening after initial yielding and the ease of crack formation and growth in thermoplastics adversely affect the usual correlation between drawability and the low strain plastic behavior of the material. They carried out experiments on polycarbonate (PC) and polyoxymethylene (POM). The true strain in the thickness direction was 0 within the experimental error in the tensile testing on biaxially rolled POM which gives an unlimited value of plastic strain ratio,  $R$ . As in previous investigations,<sup>(7)</sup> they also found that the average  $R$  value increases with the imposed tensile strain making it difficult to know which  $R$  value to use. For the polymers investigated, since they exhibit a strong work hardening in plane strain deformation which is predominant in deep drawing, the authors contend that any theory for force prediction must incorporate this work hardening. The authors believe that it is preferable to use experimental data directly instead of assuming any form of stress-strain curve. Citing the unlimited value of

R for POM, they suggest that it is impossible to calculate deep drawing forces from tensile yield stress and R value. Better agreement between prediction and experimental data was found for POM and isotropic PC with their method of calculating deep drawing forces from plane strain compression data. The agreement was not good for biaxially rolled PC. The authors feel that to predict LDR additional piece of information required is the engineering fracture stress of the material in plane strain tension that occurs in the cup wall. According to them, this can be obtained directly on the basis of one deep drawing test.

#### New Processes and Applications

Naitove<sup>(10)</sup> has described 'stretch forming' developed by Shell Oil Company. The process is a true stretching operation as discussed in the section on Principle and General Comments. The optimum temperature range specified for polypropylene is  $320 \pm 10^{\circ}\text{F}$  ( $160 \pm 6^{\circ}\text{C}$ ). In this range, the material is not tacky and shows little springback. The total part-to-part cycle time ranges from about 4 sec for smaller containers to 1/2 min for large, heavy-walled parts. The process has apparently been commercially used by one company for several years. During forming, the blank perimeter is gripped under a pressure of about 1000 psi (6.9 MPa) and the central punch applies a pressure of about 500 to 1000 psi (3.4 to 6.9 MPa) while moving at a speed of 6 to 12 ips (0.15 to 0.30 m/s). At the bottom of the stroke, air is blown through the punch tip to force the plastic wall against the mold cavity and to aid in withdrawing the punch. The side wall maintains a constant thickness despite the progressive reduction in thickness. Strong orientation results with the tensile yield strength for polypropylene increasing to 30,000 psi (207 MPa) in the vertical direction. The impact strength increases by about a factor of 10 in comparison with injection molded or blow molded parts. The process has been applied to making a wide range of parts such as a quart oil can weighing about 1 oz (0.038 kg) with an OD of 4 in. (0.10 m),



height of 5.5 in. (0.14 m), and wall thickness of 0.02 in. (0.0005 m); and a 5-gallon (0.019 m<sup>3</sup>) heavy-duty shipping container weighing 1.8 lb (0.82 kg) with an OD of 12 in. (0.30 m), height of 13 in. (0.33 m), and wall thickness of 0.08 in. (0.002 m). A prototype production of a 20-gallon (0.076 m<sup>3</sup>) washing machine tub has been mentioned. The tonnage requirement with the process is less than 10% that for injection molding, and tooling cost is only about 10 to 15% that for injection molding.

Schaper and Cronenberger<sup>(11)</sup> have described a high-speed solid-phase forming method developed and patented by Dow Chemical Company. The scrapless forming process (SFP) starts with square blanks which are lubricated and heated. In the first station of the forming machine the blank is preformed into a circular shape with a lip, and then in the next station the lip is clamped tightly and the center portion of the preform is stretched to form the cup. The main advantages of the process are claimed to be elimination of trim scrap, improvement in the physical properties due to biaxial orientation, and simplicity of forming with short cycle times. For styrenic polymers, the preheating temperature is in the range of 230° to 280°F (110° to 130°C). For crystalline polymers such as high-density polyethylene and polypropylene, the range is somewhat narrower but, once again, below the crystalline melting point.

The process is particularly well suited to multi-layer blanks and, with proper lubrication, the thickness of each layer as a percentage of the total thickness remains remarkably constant at all points in both the preform and the finished part. The elimination of trim scrap makes SFP attractive for fabricating multi-layer containers since cycling of scrap that includes incompatible polymers is frequently impractical. The biaxial orientation during the process improves tensile strength,

toughness and stress crack resistance. For high-density polyethylene, the increase in tensile strength can be as much as 300%. Therefore, part thickness can be reduced or expensive high-strength resins can be replaced by lower cost, high impact polystyrene for applications that require environmental stress crack resistance. Such changes have important cost-saving potential. Rubber-modified impact resins manifest greater clarity with SFP. The typical SFP cycle for high impact polystyrene is only 2 sec and for high-density polyethylene 2.5 to 3 sec.

The paper provides some cost comparison based on an 8 fluid oz ( $2.4 \times 10^{-4} \text{ m}^3$ ) capacity tub-like container 4.125 in. (0.1048 m) in diameter by 1.875 in. (0.0476 m) deep. A 9 g container made from high impact polystyrene by SFP is nearly 11% lower in cost than a thermoformed ABS part of the same weight. Further cost reduction is possible by decreasing the container wall thickness of the SFP formed part. In another comparison, when no scrap recycling is allowed, a transparent multi-layer tub can be produced by SFP from high impact polystyrene with a cost reduction of nearly 35% in comparison with a thermoformed ABS part.

The automotive industry--because of increasing emphasis on weight reduction and the quantities and sizes of the components required--will continue to be a major user of plastic stampings. The application is covered in detail in Ref. 12 with photographs of components being utilized or to be utilized in automobiles. References 13 to 15 provide related information on plastic stampings. It appears that a fender liner for 1977 Ford trucks and some of the passenger cars is one of the larger thermoplastic stampings in production. Two of these inner fenders\* are stamped from each sheet of unreinforced polypropylene which is then subjected to trimming. The production rate is approximately 250 parts per hour. It is implied that, through a judicious choice of processing conditions such as material temperature and dwell

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\*Each fender appears to be approximately 20 in. x 40 in. in plan area (approximately 0.5 m x 1.0 m).

time and design of molds for control of heat transfer, the problem of springback has been minimized. The material is formed below the melting point of the material using conventional metal stamping presses. The starting thickness of the sheet is approximately 80 mils (0.002 m). Ford's use of polyolefin sheet for stamped parts was estimated to be around 10,000 tons (9 Mkg) per year (according to Ref.15 dated May 1976). This is the largest known usage of unreinforced thermoplastic stampings made by solid-phase forming. Other polymers that have been mentioned in connection with stamping include ABS and nylon.

In the past, usage of glass-reinforced polypropylene sheet (such as GRTL's Azdel) has been reported in the automotive industry. Unlike the above-mentioned stamping, Azdel is formed at a temperature above the melting point of the resin and is thus not solid-phase forming. However, the technique employed is similar to metal stamping. The total usage of plastic stampings<sup>(15)</sup> was 12,000 tons (10.8 Mkg) per year (most of it polypropylene) and was expected to rise to at least 60,000 tons (54 Mkg) per year by 1981--with nylon, polyethylene, thermoplastic polyester being the common polymers. Various nonautomotive applications of the plastic stampings have also been mentioned such as a motor shroud and air baffle for the Lawn-Boy mower. The shroud, measuring roughly 16 in. x 12 in. (0.41 m x 0.30 m) and weighing about 1 lb (0.45 kg), is stamped on a 250-ton (2.2 MN) hydraulic press in a 27 sec cycle using 100 mil (0.0025 m) sheet. Conventional molding of such large thin parts is difficult.

It is clear that the automotive industry has made a major commitment to usage of plastic stampings both from unreinforced and reinforced materials. For example,<sup>(14)</sup> a 5-year development program preceded Ford's usage of the polypropylene stampings. It is, therefore, safe to predict that the usage of plastic stampings will grow at a fairly rapid pace in the years to come.

## FORGING

### Principle and General Comments

Forging is a process in which a preform or blank is subjected to bulk deformation by application of force with constraining dies which are generally made from steel. Apart from the simple case of upsetting between flat platens, a forging is characterized by a substantial degree of variation in its section thickness and often a significant degree of lack of symmetry. Whereas the stamping operations are normally carried out on relatively thin sheets with thickness of the order of 0.08 in. (0.002 m) and rarely in excess of 0.2 in. (0.005 m), forgings are often made from billets which may be several inches thick. Although materials such as polypropylene can be cold forged in theory, the high degree of springback makes warm forging essential for most thermoplastics.

The main advantages of the forging process are a high utilization of material and a substantial reduction in machining costs since machining an oversize billet is often the only alternative to making a complex part by forging. As in the case of stamping, generally some property improvement occurs in forging--mainly in the strength with sometimes some reduction in the ductility of the material. Also, forging allows production of complex shapes in materials such as ultra high molecular weight polyethylene (UHMWPE) which are practically impossible to injection mold. Even in materials that can normally be processed by injection molding, forging allows production of much thicker components with variation in section thickness at a relatively fast production rate using equipment and molds substantially lower in cost than are required for injection molding. As a general rule, forging should be considered only when the component is so thick or has such variation in section thickness that it is beyond the capability of satisfactory and economic production by injection molding.

Springback remains the major problem in forging. The severity of the problem is much more than for stamping because

of the usual lack of symmetry and variation in section thickness in forging. The crystalline materials are normally easier to forge since they show less springback. Forging of amorphous materials is extremely difficult, and especially for materials such as polycarbonate with an incorrect choice of the material and die temperatures and other process variables, a cylindrical billet may show practically complete springback to the original billet dimensions when the dies are opened after forging. Depending on the particular grade and the quality of the starting billet, cracking is more of a problem in a material such as rigid PVC. PP is the easiest material to forge in terms of capability for large deformation and low springback. UHMWPE and HDPE are also fairly easy to forge, with the latter showing higher springback.

#### Basic Investigations

In comparison with the work carried out on stamping, very little new work has been reported on forging in the past several years. This section is based mainly on the research conducted at IIT Research Institute.

Generally, the amorphous materials show a greater degree of springback than crystalline materials. The dimensional changes occur on removal of the forging from the dies and its storage at the ambient temperature. Further dimensional changes occur if the forging is subjected to higher temperatures. It is important to understand these phenomena better so that forging dimensions will be stable during usage.

Simple upsetting or compression tests were carried out on cylindrical billets of HDPE and PP to compare their springback characteristics. The initial billet dimensions were 1 in. (0.025 m) diameter x 1.25 in. (0.032 m) height. Billets heated to 225°F (107°C) were subjected to 60% and 80% reduction in height corresponding to die closure of 0.5 in. (0.013 m) and 0.25 in. (0.006 m), respectively, between flat platens. The

heights of the forgings were recorded immediately on removal from the platens as well as after storage at ambient temperature for up to several days. It was observed that essentially stable dimensions are reached in only about 5 min with the HDPE showing a greater springback (larger final height) than PP. Subsequently, the billets were heated to 225°F (107°C) for several hours and their dimensions were periodically checked. The data are shown in Fig. 6. Note that most of the dimensional change takes place within the first 8 hr. Then the forgings should be quite stable for any usage below the temperature of the thermal treatment.

For a given configuration of the die cavity, the degree of deformation imposed on a preform depends on its dimensions and, therefore, the springback varies considerably depending on the preform dimensions. The complexity of the component geometry and variation in its section thickness can cause large differences between forging and die cavity dimensions. Generally, the dimensional fidelity is better the higher the forging temperature and the longer the duration of load. However, much remains to be investigated concerning optimization of the preform geometry and process parameters.

In IITRI's work, forgings were made to the configuration shown schematically in Fig. 7. These are the dimensions of the forging at die closure, i.e., the ideal forging dimensions. The forgings were made using the die set shown in Fig. 8. Notice the simplicity of the die set in comparison with a complex injection molding mold that would be required for a component of the size and shape in Fig. 7. The die set is of completely closed type, i.e., it does not generate any flash on the forging. Table 2 shows satisfactory processing conditions and the dimensions of the resulting forgings for a series of materials. These data will give some idea of the optimum processing conditions for these materials. Further, the material temperatures can be compared to the transition temperatures in Table 1. Note also the large differences in the actual dimensions as against the ideal dimensions in Fig. 7.

Materials such as PP, HDPE, and UHMWPE have large ductility and can be subjected to much deformation even at low temperatures without cracking. On the other hand, engineering plastics such as acetal need much closer control over processing conditions. As shown in Fig. 9, too low a forging temperature can cause catastrophic breakage in an acetal forging. Although generally not recognized, the deformation speed is also important in forging of low ductility plastics for both the amorphous and crystalline types. A recent patent addresses this effect.<sup>(16)</sup> Nylons have good ductility but need higher temperatures and loads for forging. They may also show some surface degradation. Amorphous materials such as ABS, PVC, and polycarbonate can also be forged but need very close control over processing conditions. Unlike for crystalline materials, tooling temperature is very important for the amorphous plastics.

Most crystalline polymers are forged in the solid phase, i.e., below their melting point. However, UHMWPE does not become fluid even when heated to a fairly high temperature in excess of the melting point. During forging, a somewhat higher temperature above the melting point leads to dimensions closer to the ideal forging dimensions (Table 2) while still retaining many of the advantages of the forging process. The billet can be handled readily even when the temperature is approximately 75°F above the nominal melting point of 275°F. This is similar to the hot stamping of the glass-reinforced PP discussed earlier where the reinforcement allows handling above the melting point of the matrix resin. The point is that one should not be too restrictive in selecting the processing temperature just to call the process solid-phase forming. Instead, the most appropriate temperature should be selected for the application and material concerned.

## New Processes and Applications

Several UHMWPE forgings have already found commercial applications such as the snowmobile sprockets shown in Ref. 2. Polypropylene forgings are also in use. A limited amount of new work has been reported concerning processing techniques. For example, billets can be produced<sup>(17)</sup> by filling a series of interconnected molds contained in a cooling chamber of sufficient length to effect complete solidification. An accumulator system fills the molds and maintains pressure during solidification. The shrinkage that occurs as the melt solidifies is compensated for by molten polymer flowing through the interconnected mold system under pressure. It is mentioned that a prototype machine can produce billets from 200 to 700 cm<sup>3</sup> in any thermally stable polymer which can be extruded in a conventional extruder.

Much of the earlier work was on forging of unreinforced billets. Recently billets of glass-reinforced material have also been made and subjected to forging.<sup>(13)</sup> There is also mention of a new process that combines injection molding and forging.<sup>(18)</sup>

The forging process has much potential for application for thick and relatively complex components. However, as far as is known, no large potential user has made any major commitment and much work remains yet to be performed. Without such major commitment and additional work, the growth in the usage of plastic forging is likely to be quite slow for some time to come. Smaller quantities of forgings will find applications in special cases where the economic advantages of the process or its unique capabilities are unchallenged.

## OTHER SOLID-PHASE FORMING PROCESSES

A variety of forming techniques have been mentioned in literature as reviewed in Ref. 1. By and large, these are variations of the stamping and forging techniques discussed



in this paper. Little additional work has been reported on these different variations. An example of work that can be considered to be similar to solid-phase processes is high-pressure nonisothermal processing (HIPRENIP),<sup>(19)</sup> This is essentially a compression molding technique in which the temperature is only slightly above the melting point and the compression molding pressure is very high. For high-density polyethylene, usage of 80,000 psi (552 MPa) pressure led to components with significantly higher rigidity than similar conventionally processed material. Very likely the technique could be applied to other polymeric materials.

#### CONCLUDING REMARKS

In the past five years, a significant amount of work has been carried out on stamping and major applications of plastic stampings can be found in the automotive industry. Some applications have found their way into other industries also. The usage of plastic stampings is likely to grow at a rapid pace. Comparable growth in the application of plastic forgings can occur only after additional research work and serious commitment by one or more large potential users.

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Table 1  
TRANSITION TEMPERATURES FOR SOME  
COMMON THERMOPLASTICS<sup>(3)</sup>

Crystalline Polymer	Melting Point, °C
Acetal Homopolymer	181
PTFE	327
Nylon 6/6	265
HDPE	120-140
UHMWPE	120-135
PP	176
Amorphous Polymer	Glass Transition Temp., °C
High Impact ABS	100-110
Polycarbonate	150
PS	100
Rigid PVC	75-105

Table 2  
FORGING DATA FOR VARIOUS MATERIALS

Material	Preform	Material Temp., °C	Die Temp., °C	Press Speed, mm/s	Load Duration, s	Forging Size <sup>a</sup> Dia. x Ht., mm
PP	A <sup>b</sup>	149	66	Creep <sup>c</sup>	120	123.4 x 45.2
PP	A	149	66	Creep	30	118.6 x 45.0
PP	A	RT <sup>d</sup>	RT	Creep	180	118.1 x 42.7
Acetal	A	163	135	Creep	30	122.7 x 42.4
UHMWPE	B	177	RT	2.5	120	124.5 x 46.7
UHMWPE	B	135	RT	2.5	120	124.5 x 43.2
HDPE	B	121	66	2.5	120	121.9 x 44.5
Rigid PVC	B	107	66	2.5	120	124.5 x 44.5
Rigid PVC	B	107	RT	2.5	180	124.5 x 47.2
ABS	B	102	66	2.5	120	125.8 x 44.5

<sup>a</sup>Ideal forging dimensions are 127 mm dia. x 47.6 mm. maximum height as shown in Figure 7.

<sup>b</sup>Preform A was 88.9 mm dia. x 44.5 mm thick and B was 101.6 mm dia. x 33.8 mm thick.

<sup>c</sup>'Creep' refers to very slow press speed.

<sup>d</sup>'RT' is room temperature.

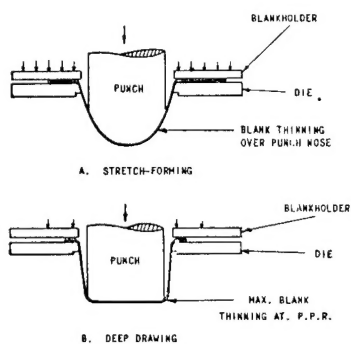


Figure 1  
Types of Deformation

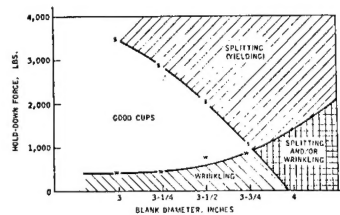


Figure 2  
Results of Limiting Draw Ratio Experiment<sup>(5)</sup>

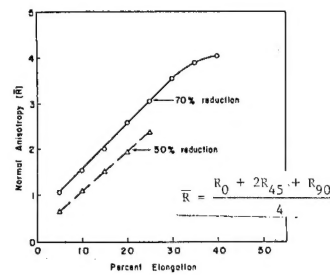


Figure 4  
Normal Anisotropy vs. Percent Elongation of Tensile Specimen for Biaxially Rolled PVC Polymer<sup>(7)</sup>

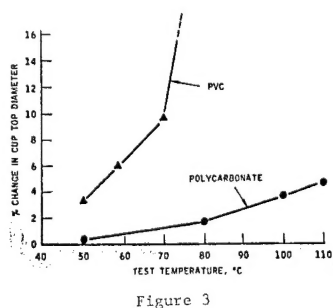


Figure 3  
Relaxation in Shape vs. Temperature with Cold Formed PVC and PC Cups<sup>(5)</sup>

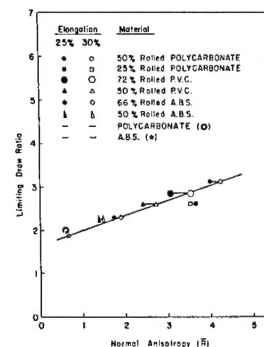
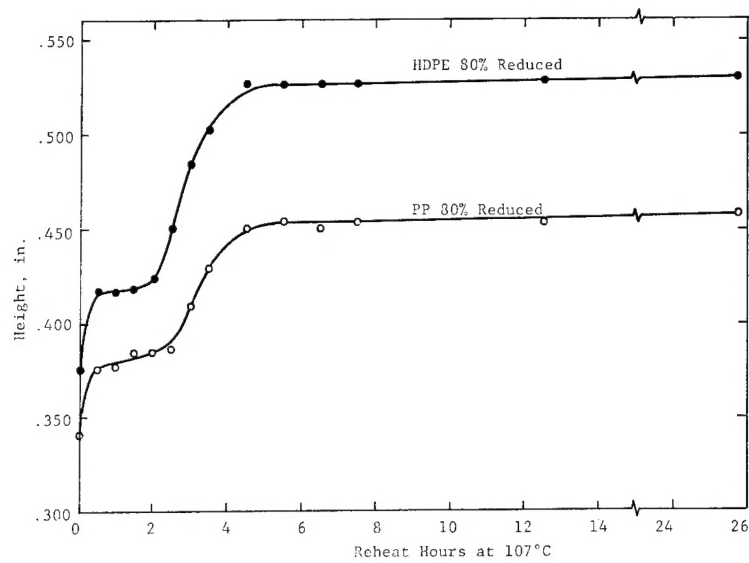


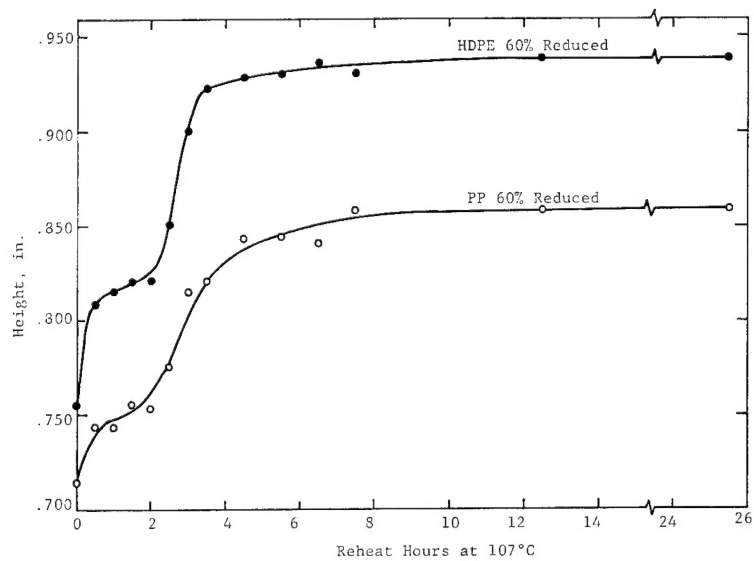
Figure 5  
Limiting Draw Ratio vs. Normal Anisotropy<sup>(7)</sup>



(a) After 60% reduction in upsetting

Figure 6

Height Changes on Cylindrical Pieces Heated to 107°C, Upset and Then Reheated at 107°C



(b) After 80% reduction in upsetting

Figure 6 (cont.)

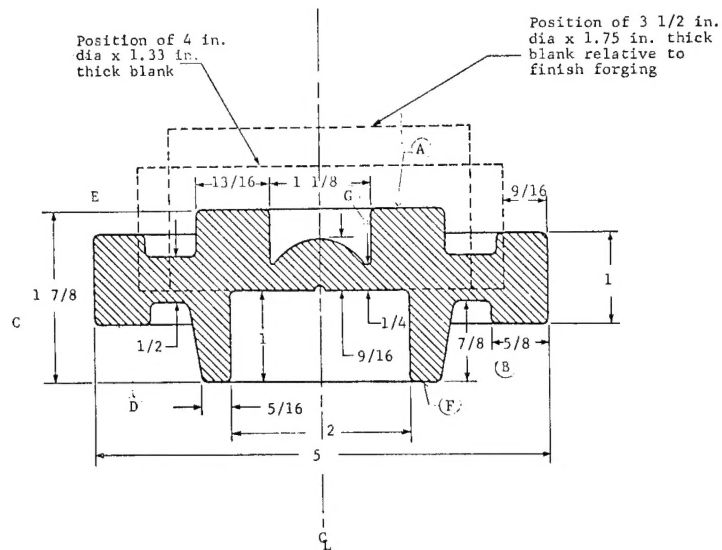
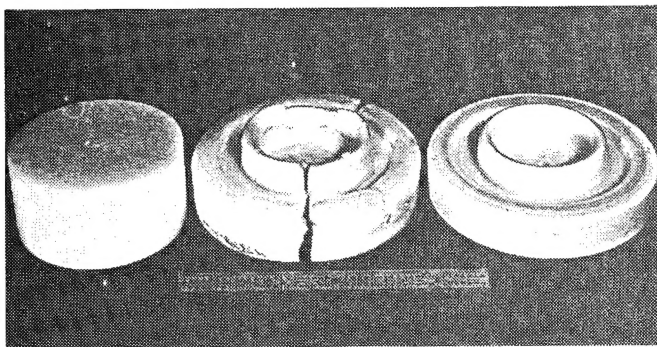


Figure 7

Forging Dimensions Inside Closed Dies and Relative Positions of Two Sizes of Blank (For large 5 in. diameter forging)



(a) (b) (c)

Figure 9

Criticality of Forging Temperature for Acetal:  
Blank (a) and Forgings Made at (b) 121°C and (c) 163°C

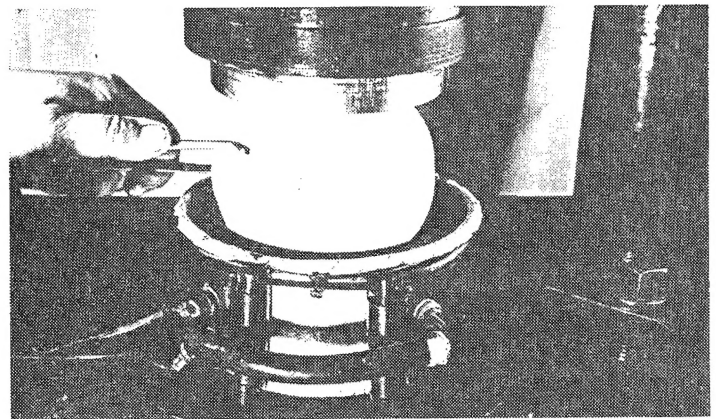


Figure 8

Forging Being Removed from the Dies  
(Notice the small size of the dies.)

Fields To Be Put on

Master Cassette

(8) u

(20) u

(28) u

(43) 1

(33) 1, 24

(11) 27 Sep 77

(22)

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